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On the application of single parameter aging to glass equilibration

Valerio Di Lisio  ; Daniele Cangialosi  



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Valerio Di Lisio¹ and Daniele Cangialosi^{1,2,a)}

AFFILIATIONS

¹ Centro de Física de Materiales (CSIC-UPV/EHU), Paseo Manuel de Lardizabal 5, 20018 San Sebastián, Spain

² Donostia International Physics Center, Paseo Manuel de Lardizabal 4, 20018 San Sebastián, Spain

^{a)} Author to whom correspondence should be addressed: daniele.cangialosi@ehu.eus

ABSTRACT

The kinetics of physical aging in glasses is typically described by models accounting for the material time concept, which, via the time dependent relaxation time—the material “inner clock”—linearizes the response. Although this description has been shown to be successful for relatively small temperature jumps, typically close to the glass transition temperature, T_g , its validity for large jumps is a matter of open debate. Here, we implement the material time concept within the single parameter aging model with a single time scale, that is, the one of the main α relaxation, and apply it to isothermal aging data after both temperature down- and up-temperature jumps. Our results show that a single parameter falls short of capturing the entire phenomenology of physical aging. Specifically, a single parameter can either describe aging after temperature up-jumps and relatively small down-jumps or capture the entire range of aging kinetics after temperature down-jumps while falling short of predicting aging after up-jumps. This outcome highlights the necessity of incorporating multiple time scales in the description of physical aging.

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I. INTRODUCTION

Liquids can generally be supercooled below their melting temperature, provided that a cooling rate large enough to avoid crystallization is employed. On further cooling, the supercooled liquid transforms into a glass at a temperature depending on the applied cooling rate, commonly addressed as T_g . The so-formed glass is in non-equilibrium and, therefore, its free energy decreases with time until it equals that of the supercooled liquid, a phenomenon commonly addressed as physical aging^{1,2} or structural recovery.³

The mechanism underlying the transformation of the supercooled liquid into a glass and the subsequent time dependent evolution in the glassy state is a matter of intense debate.² While the role of the main α relaxation, with super-Arrhenius temperature dependence, has traditionally been emphasized,⁴ in recent years, indications that in some conditions other mechanisms may play a role in both vitrification and physical aging have been provided. Specifically, the direct relation between vitrification kinetics and α relaxation time, τ_α , over a wide range of conditions, while valid in several systems,^{5,6} is broken in both bulk⁷ and confined glasses.^{8,9}

In the case of physical aging, the most prominent feature of the presence of different mechanisms of equilibration is the finding of multiple plateaus toward glass equilibration when experiments are carried out over a wide temperature and time range.^{10–13}

A hallmark of physical aging is its nonlinear character: as the thermodynamic state of the glass evolves over time, the structural relaxation time, τ_α , changes accordingly. The nonlinear nature of physical aging is described by the concept of material time, ξ , long ago introduced by Narayanaswamy,¹⁴ which involves a linearization of the absolute aging time, t , via the clock rate, that is, the inverse of τ_α ,^{14–17}

$$d\xi = \tau^{-1} dt. \quad (1)$$

Within the material time approach, the non-linear response underlying the recovery of equilibrium can be fully described by the linear response, that is, the response to small perturbations fulfilling the fluctuation–dissipation theorem.

The material time concept has recently been incorporated into the single-parameter aging (SPA) model. Within this framework, the

sole fitting parameter is the proportionality coefficient linking variations in $\log \tau_\alpha$ to changes in the thermodynamic state of the glass, such as its enthalpy or volume. If applied to aging after relatively small jumps, that is, in the neighborhood of T_g , the SPA model has been shown to provide an excellent prediction of the aging time evolution of a given observable.^{15–18} In other words, it is possible to predict non-linear equilibrium recovery kinetics from the glass response in the linear regime.^{17,19}

Recently, within the SPA model, the material time concept, with a single time scale (that of the α relaxation), has been extended to describe the evolution of the enthalpic state of different small molecule glasses during physical aging after temperature jumps of more than 20 K.²⁰ In the approach used, the single parameter was chosen *ad hoc* to fit data at all aging temperatures, although the physical soundness of such a parameter remained elusive. Specifically, it was not clear what the order of variation of $\log \tau_\alpha$ calculated from its relation with the glass enthalpy via the fitted single parameter was, nor how the linearized response via the single parameter compares with the linear response taken from independent measurements.¹⁷ Furthermore, the ability of the SPA approach to catch aging after large temperature jumps was severely questioned when the connection of τ_α to the glass thermodynamic state was established via the density scaling framework,²¹ recently tested in non-equilibrium conditions. In this case,²² the aging-time dependence of τ_α is obtained with no adjustable (fitting) parameters. In this way, the prediction of enthalpy evolution, while accurate for small temperature jumps (up to ~ 6 K), was considerably slower than the experimental one in the initial stages of aging. This failure was attributed to the inability of the α relaxation alone to describe physical aging when this is investigated considerably below T_g , as previously discussed.

In the present study, we address the issue of the suitability of the SPA model with a single α time scale extending the enthalpy relaxation set of data on small molecule glasses to aging after temperature up-jumps. We show that the single fitting parameter extracted from small down and up-jumps to the same temperature falls short in predicting the kinetics of physical aging after large temperature down jumps. Likewise, if fitting is carried out for the whole range of temperature down-jump aging experiments, the so-obtained single parameter is unable to capture the kinetics of aging experiments after temperature up-jumps.

II. EXPERIMENTAL

o-Cresolphthalein dimethyl ether (KDE, $\geq 98\%$ purity), phenolphthalein dimethyl ether (PDE, $\geq 98\%$ purity), and 1,1-bis(4-methoxyphenyl)cyclohexane (BMMPC, $\geq 98\%$ purity) were supplied by Polymer Source. *o*-Terphenyl (OTP, $\geq 99\%$ purity) was purchased from Sigma-Aldrich, while BMPC (also known as bisphenol-C dimethyl ether) was provided by the Max Planck Institute for Polymer Research (Mainz, Germany). All compounds were used without further purification.

Fast Scanning Calorimetry (FSC) measurements were carried out using a Mettler-Toledo Flash DSC 1, equipped with an intra-cooler and operating over a temperature range of 173–723 K. The sample chamber was continuously purged with dry nitrogen at a flow rate of 20 ml/min. Samples were prepared by directly depositing the material (with a mass of ~ 100 ng) onto a Mettler-Toledo UFS1 chip. The deposited specimens had an equivalent diameter of about

200 μm ; considering their mass (~ 100 ng) and material densities in the range of 1–1.3 g/cm³, this corresponds to an estimated thickness on the order of 10 μm .

Isothermal aging experiments were preceded by heating and cooling scans at a rate of 1000 K/s in a temperature range between 173 K and $T_m + 10$ K, where T_m is the melting temperature of the compound. Samples were subsequently cooled to the first target aging temperature, $T_{a,1}$, and maintained under isothermal conditions for a sufficiently long time to attain equilibrium. Subsequently, a temperature up-jump was applied to the samples to the second target aging temperature, $T_{a,2}$, where isothermal annealing at different times was carried out.

Following this thermal treatment, the samples were rapidly cooled to 173 K, and a subsequent heating scan was acquired. Before determining the time- and temperature-dependent fictive temperature, $T_f(T_a, t_a)$, the glassy and liquid heat-flow signals from each experiment were aligned with the corresponding unaged reference scan using the VITRIFAST method. Recently introduced by Abate *et al.*,²³ this optimization procedure minimizes the mean squared error between aged and unaged scans in both the glassy and liquid regions.

III. SINGLE PARAMETER AGING MODEL

The SPA model is inspired by the seminal study of Narayanaswamy,¹⁴ who introduced the concept of material time to account for the aging time/thermodynamic state dependence of the relaxation time. To this aim, the SPA model makes the simplest connection between τ and any property, P , sensitive to the glass thermodynamic state,^{15–17}

$$\log \tau(t) - \log \tau_{eq} = \Lambda(P(t) - P_{eq}), \quad (2)$$

where Λ is the single parameter connecting the aging time dependent $\tau(t)$ with $P(t)$. In the present study, as we monitor the aging time dependent enthalpic state of the glass, we identify P with the fictive temperature, T_f . Introduced long ago by Tool,²⁴ it is defined as the temperature at which the glass line drawn from the glass enthalpic state intersects with the supercooled liquid line. T_f is therefore a measure of how deep in the energy landscape a glass is.

Starting from the time dependent T_f and its values at the beginning, T_{f0} , and the end of aging, that is, the aging temperature, T_a , we define the normalized relaxation function, varying between unity at the beginning of aging and zero once aging is completed,

$$R(t) = \frac{T_f(t) - T_a}{T_{f0} - T_a}. \quad (3)$$

To model the physical aging behavior of a given glass, knowledge of the normalized relaxation function at a reference aging temperature, $R_1(t)$, is required. This function can be obtained by interpolating or fitting the experimental aging data collected at the reference temperature with an appropriate model function. Within the description based on the material time, the non-linear response after a generic temperature jump can be linearized if the absolute time is replaced by the material time. In this way, regardless of the temperature, the normalized relaxation function is uniquely determined by the material time, i.e., $R_1(\xi) = R_2(\xi)$. Consequently, the

normalized relaxation function corresponding to an arbitrary temperature jump can be derived from the relaxation function measured at the reference temperature.¹⁷

The equality of all relaxation functions independently of the temperature jump also applies to the first derivatives,

$$\frac{dR_1(\xi)}{d\xi} = \frac{dR_2(\xi)}{d\xi}. \quad (4)$$

The latter equation can be rewritten in terms of the absolute time using Eq. (1),

$$\frac{dR_1(t_1^*)}{dt_1^*} \tau_1(t_1^*) = \frac{dR_2(t_2^*)}{dt_2^*} \tau_2(t_2^*). \quad (5)$$

We now introduce the connection of τ to the glass thermodynamic via the single parameter, Λ ,

$$\frac{dR_1(t_1^*)}{dt_1^*} \exp(\Lambda R_1(t) \Delta T_{f1}) = \frac{dR_2(t_2^*)}{dt_2^*} \exp(\Lambda R_2(t) \Delta T_{f2}), \quad (6)$$

where ΔT_{f1} and ΔT_{f2} are the total changes in the fictive temperature after a temperature jump. If we consider identical variations in the normalized relaxation function, $dR_1(t_1^*) = dR_2(t_2^*)$, the corresponding time increment for aging at the target temperature can be obtained by rearranging Eq. (6),

$$dt_2^* = \frac{\tau_{eq,2}}{\tau_{eq,1}} \exp[R_1(t_1^*) \Lambda (\Delta T_{f2} - \Delta T_{f1})] dt_1^*. \quad (7)$$

Which can be integrated,

$$t_2 = \int_0^{t_2} dt_2^* = \frac{\tau_{eq,2}}{\tau_{eq,1}} \int_0^{t_1} \exp[R_1(t_1^*) \Lambda (\Delta T_{f2} - \Delta T_{f1})] dt_1^*. \quad (8)$$

Hence, it is possible to predict the aging kinetics in terms of the relaxation function at any temperature, once the reference relaxation function after a given temperature jump and the single fitting parameter, Λ , are known.

For temperature jumps at the same temperature: $\tau_{eq,2} = \tau_{eq,1}$, which simplifies Eq. (8). Hence, several studies have made use of down- and up-jumps to the same temperature to extract the parameter Λ , which was later used to predict aging kinetics after a variety of thermal protocols.¹⁷ Alternatively, as performed in Ref. 20, it is possible to consider a set of aging experiments after different temperature jumps and obtain the Λ parameter that best fits the aging kinetics.

IV. RESULTS

Figure 1 shows the heating scans after the two-step aging protocols described in Sec. II for all investigated glasses. The figure presents the temperature dependent heat flow, HF , which is proportional to the specific heat: $c_p \sim HF$. The hallmark of the aging

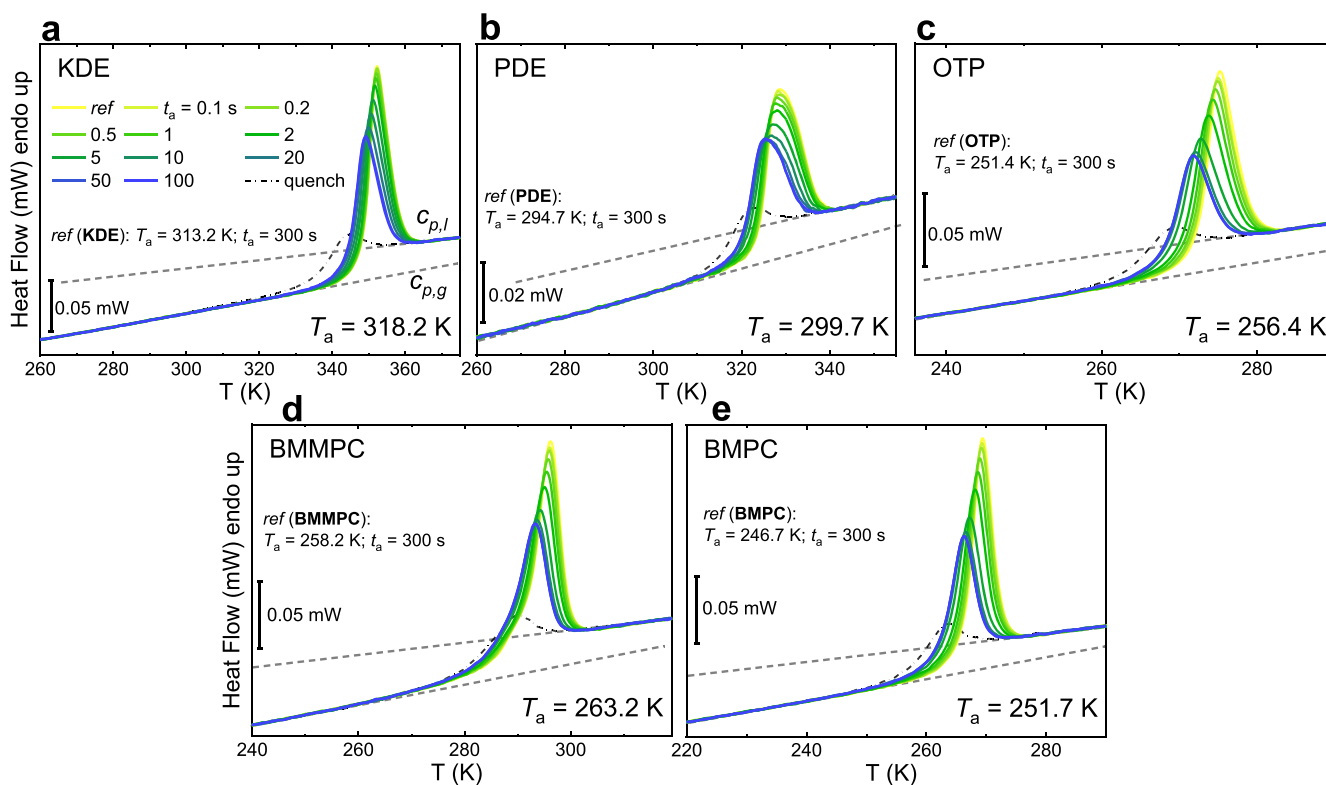


FIG. 1. Aging time dependent heat flow scans for temperature up-jump experiments at the indicated temperature. Glasses were previously equilibrated for 300 s at the indicated lower temperature.

time evolution of heat flow scans is the reduction of the endothermic overshoot, implying that the enthalpy of the glass is increasing. This is expected and in line with previous reports,^{25,26} considering that the glasses were previously aged at a lower temperature and the equilibrium enthalpy after the temperature up-jump resides at a higher enthalpic state.

From the relation between specific heat and enthalpy, $c_p = \left(\frac{\partial H}{\partial T}\right)_p$, the area enclosed between the aged and unaged scans provides the amount of enthalpy relaxed during physical aging and, therefore, the T_f variation. The latter can be determined using the Moynihan method,²⁷

$$\int_{T_f}^{T_1} (c_{p,l} - c_{p,g}) dT = \int_{T_2}^{T_1} (c_p - c_{p,g}) dT, \quad (9)$$

where c_p is the specific heat of the sample, while $c_{p,l}$ and $c_{p,g}$ are the liquid and glass baselines, respectively, obtained by linear fitting (see dashed lines in Fig. 1, where this is performed on H_f given its proportionality to c_p). The temperatures T_1 and T_2 are selected to be well above and below T_g , respectively. The Moynihan method allows determining the T_f at the beginning of aging, that is, just after cooling at 1000 K s^{-1} . The latter roughly corresponds to the T_g at that cooling rate. These are: 330.5 K for KDE; 306.5 K for PDE; 263 K for OTP; 277 K for BMMPC; and 259 K for BMPC.

The time dependent T_f (shown in Fig. 1 of the [supplementary material](#)) allows determining relaxation functions, $R(t)$, via Eq. (3). The latter is shown in Fig. 2 as a function of the aging time for all investigated systems after down- and up-jumps at the same temperature. As can be observed, $R(t)$ after temperature up-jumps exhibits markedly compressed behavior, whereas the opposite is observed after temperature down-jumps, which rather display a stretched approach toward equilibrium. This is a common feature in this kind of experiment, commonly addressed as asymmetry of approach to equilibrium^{3,11} and explained straightforwardly as resulting from the non-linearity of physical aging, as discussed in Sec. III.

Relaxation functions from temperature down- and up-jumps at the same temperature can be used to obtain the Λ parameter of the SPA model. Specifically, from the reference relaxation function in the temperature down-jump experiment, the relaxation function after the temperature up-jump can be generated via Eq. (8) with $\tau_{eq,2} = \tau_{eq,1}$. The continuous lines in Fig. 2 are the best fits to temperature up-jumps via the indicated Λ value.

The latter can be used to predict the relaxation function after the temperature down-jumps. The outcome of this analysis is reported in Fig. 3, where the predictions with the Λ parameters obtained from down- and up-jumps at the same temperature are reported as continuous lines and confronted with previously published aging data.²² Observation of the figure indicates that the predictions are accurate over the entire aging time range for

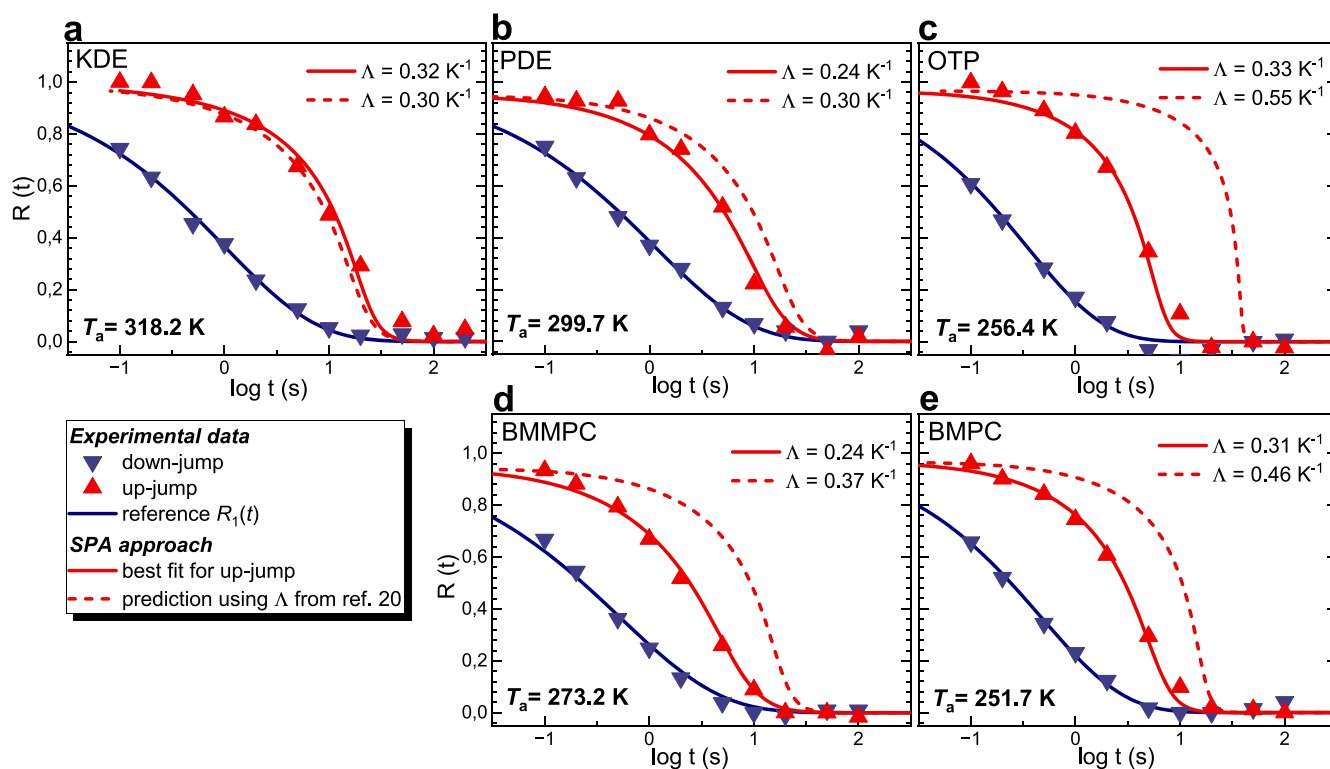


FIG. 2. $R(t)$ of up-jumps and down-jumps at the reference T_a for all investigated systems. The continuous blue lines for the temperature down-jumps at T_a represent the reference relaxation functions $R_1(t)$, obtained by a KWW fit. The continuous red lines show the best SPA fit for the up-jumps, obtained by optimizing Λ in Eq. (8). The dashed red lines are the predictions of the SPA model using Λ parameters obtained from the best fit of the series of aging experiments after temperature down-jumps.

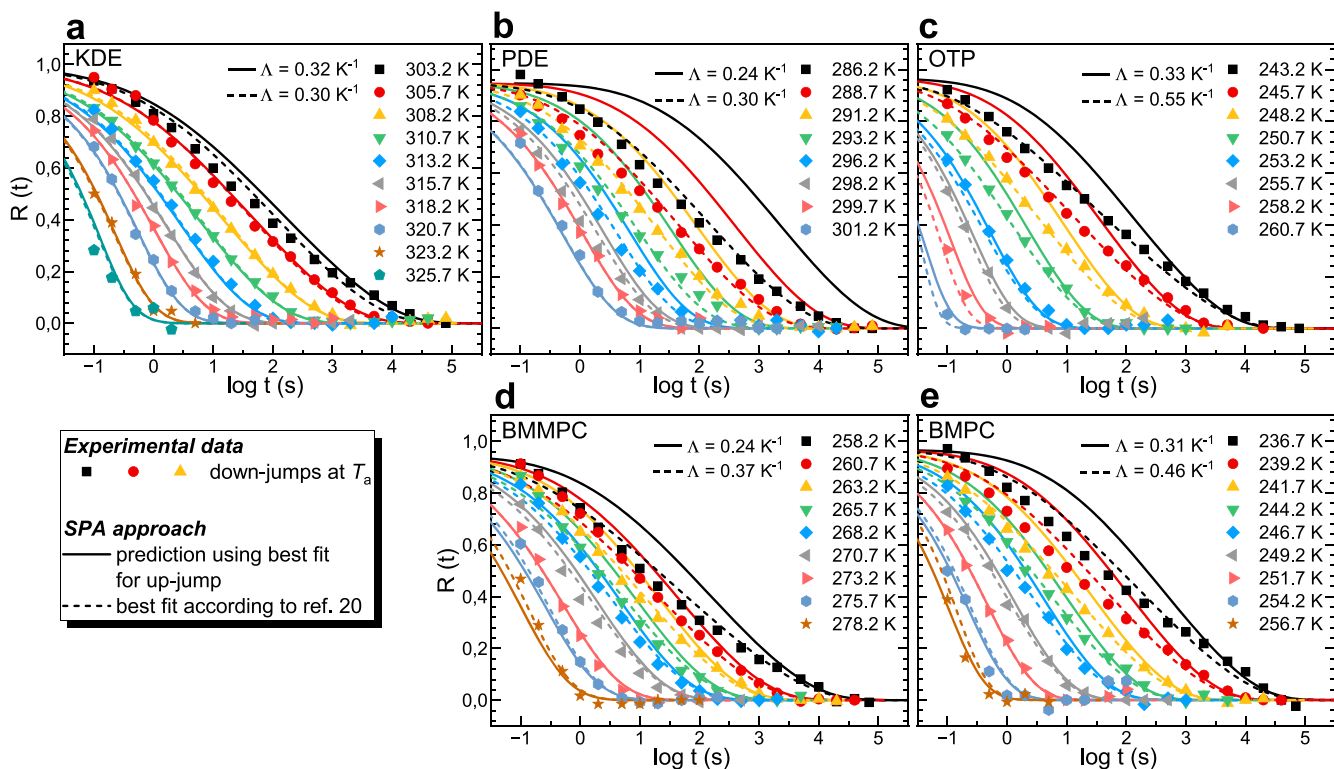


FIG. 3. SPA description for down-jump aging series results. The thick black lines are the predictions with the Λ values that best fit the up-jump series, shown in Fig. 2. The dashed lines are the best fits for the overall down-jump series using the Λ parameter obtained in temperature down-jump experiments.

down-jumps until an aging temperature that depends on the glass. In contrast, at the lowest aging temperatures, the same predictions fall short of describing enthalpy relaxation in the short-to-medium aging time range and are only able to catch the latter stages of aging. For PDE, OTP, BMMPC, and BMPC, deviations are observed at the aging temperatures relatively close to T_g , typically less than ~ 10 K. In contrast, for KDE, deviations from the predicted aging behavior are observed only at the lowest aging temperature.

An alternative approach consists of determining the value of the parameter Λ that provides the best fit to the aging kinetics obtained from the complete series of temperature down-jumps, as performed recently.²⁰ The dashed lines and the reported Λ parameters in Fig. 3 represent the results of this analysis. The so-obtained Λ parameters can be used to predict aging kinetics after the temperature up-jumps of the present study. These predictions are shown as dashed lines in Fig. 2. The prediction appears to reasonably capture equilibration kinetics after the temperature up-jump in KDE. However, it falls short of predicting aging behavior for all other glasses after the temperature up-jumps. Specifically, the prediction via the Λ parameter obtained considering all temperature down-jumps results in predictions exhibiting considerably more compressed behavior with respect to experimental data. Furthermore, the prediction of the SPA model with the parameter obtained from temperature down-jumps experiments systematically predicts a much slower equilibrium recovery than actually observed in experiments.

V. DISCUSSION

The central aspect tackled by the present study is whether the material time approach with a single time scale, that of the α relaxation, integrated into the SPA model is able to simultaneously describe aging data over a wide range of conditions. Apart from already published temperature-down experiments,²² we have enriched the set of aging data with enthalpy relaxation results following temperature up-jumps after aging to equilibrium at a lower temperature.

The main outcome of our study is that a single parameter, Λ , can capture the phenomenology of aging if only a partial set of data on aging kinetics is considered. Specifically, a single Λ is adequate to describe aging kinetics after down- and up-jumps to the same aging temperature and relatively small temperature down-jumps (less than ~ 10 K). However, the same Λ parameter falls short of capturing the initial and intermediate stages of aging after temperature down-jumps to lower temperatures. Similarly, a single Λ parameter can describe the whole set of aging data after temperature down-jumps, but it is unable to capture the kinetics of equilibration after a temperature up-jump.

Apart from checking the ability to capture the whole phenomenology of aging, a step forward in the analysis of the kinetics of equilibration concerns the physical meaning of the Λ parameters obtained by the two different prediction approaches. Specifically, by providing the instantaneous evolution of τ via Eq. (2), the Λ

parameter allows linearizing aging data under all conditions. This is performed in Fig. 4, where the evolution of the relaxation function is shown as a function of the material time at the reference temperature, that is, the aging temperature corresponding to the smallest temperature down-jump. In the same figure, we show the linear response obtained from independent measurements. This is obtained using the Kohlrausch–Williams–Watts (KWW) function: $R(\xi) = \exp(-\xi/\tau)^{\beta_{KWW}}$, where β_{KWW} is the stretching exponent. The latter was taken from dielectric and light scattering data in proximity to the reference temperature,^{28–31} where the shape is approximately temperature invariant.²⁸ The shaded areas in Fig. 4 highlight how the relaxation function is modified considering the range of variation of β_{KWW} , mostly for those glasses where the latter exhibits different values depending on the employed technique. As can be observed, linearization of aging kinetics only approximately conforms to the least stretched bound of the linear response in KDE and PDE.³¹ In the case of the other glass formers, the single parameter obtained from the two modeling strategies is more stretched than the linear response.^{28–30} This deviation is more severe for the modeling entailing the fit of the complete series of aging kinetics after temperature down-jumps, where an unphysically compressed linear response is obtained ($\beta_{KWW} > 1$).

Apart from the intrinsic inability of a single parameter to describe aging in all conditions, it is worth considering the

physical soundness of the SPA parameters obtained in both temperature down- and up-jump experiments. Equation (2) allows determining the variation of τ during aging. For instance, this can be performed for the largest T_f variation (~ 20 K) in OTP, for which the most diverging values of Λ are obtained. This change is $\Delta \log \tau \sim 6$ and 3 for $\Lambda = 0.55$ and 0.33 K^{-1} , respectively, which, considering viscosity experiments conducted under pressure³² combined with volumetric determinations,³³ corresponds to a specific volume variation of more than 5% and 3.5% for $\Lambda = 0.55$ and 0.33 K^{-1} , respectively. In contrast, volumetric measurements on glassy and liquid OTP³³ show that the variation upon aging corresponding to a change of 20 K in T_f is of the order of 1%. Hence, the Λ values obtained from fitting aging data are largely nonphysical.

One possible interpretation of the inability of the SPA model to describe the kinetics over a wide range of conditions is that the linear relation between the change in the relaxation time and the change in a given property [see Eq. (2)] does not hold when aging after large temperature down-jumps is considered. In this case, the linear relation should be replaced by introducing higher order terms in a Taylor expansion.¹⁷ However, a different description of the aging time dependence of the relaxation time based on density scaling²¹ has been recently proposed. In that case, all parameters came from independent fits to relaxation time data at equilibrium. The outcome was analogous to that found when aging kinetics after temperature

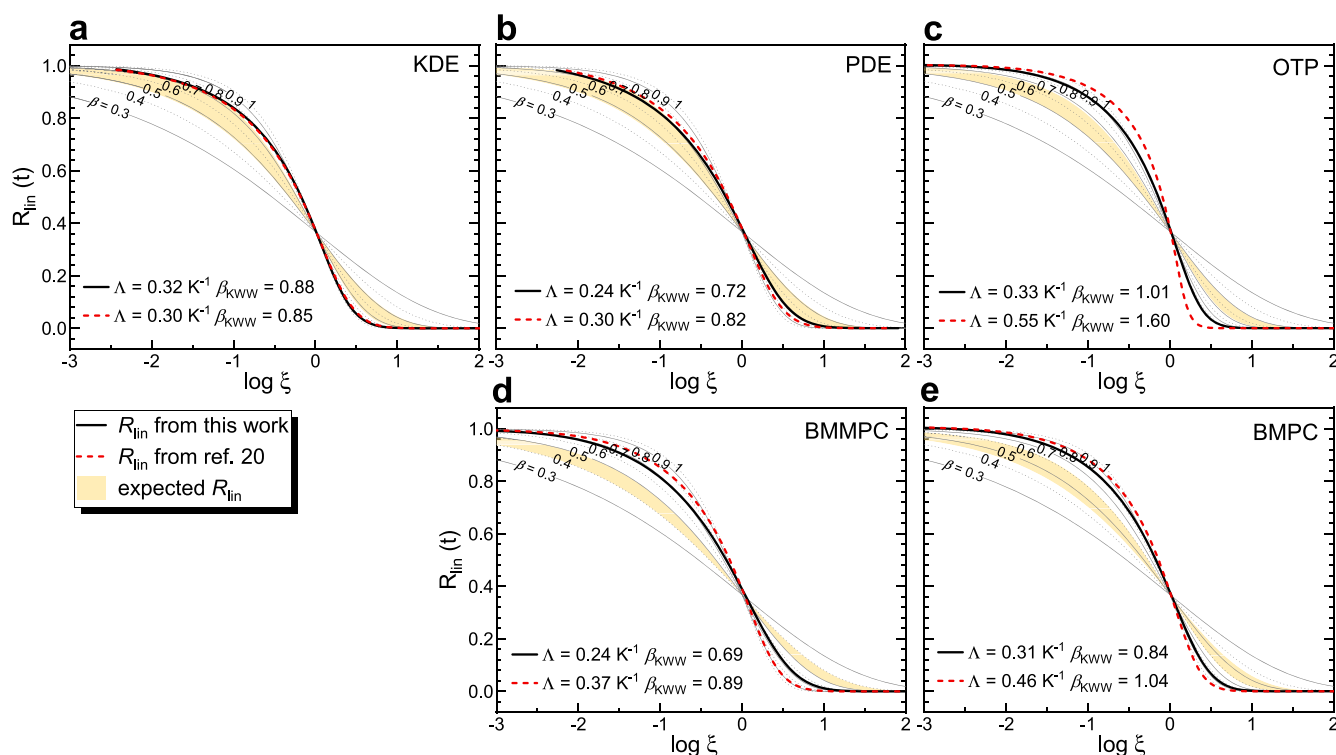


FIG. 4. Linear response of the normalized relaxation function R_{lin} as a function of the material time. The prediction with the values of Λ obtained from fitting aging results after down- and up-temperature jumps at the same temperature (black thick line) and those from the fits via the Λ parameter obtained in temperature down-jump experiments (dashed red lines) are shown together with a range of variation of the linear response (black thin lines), emphasizing the variation from experiments (shaded yellow area).

down-jumps is described from the Λ parameter obtained by fitting aging kinetics after temperature up-jumps (see continuous lines in Fig. 3).

Furthermore, there exists increasing evidence that for large temperature down-jumps the approach to equilibrium takes place in different steps, with at least one intermediate plateau. This has been found in polymers,^{11,13,34–36} chalcogenides,^{10,13,37} metallic glasses,^{12,38} and glycerol.³⁹ Indications of an incipient splitting into two steps come from observation of aging kinetics for the largest temperature down-jumps. For instance, aging at 236.7 K in BMPC exhibits evidence of this feature at $\sim 10^3$ s, an observation that warrants exploring aging at lower temperatures. The monotonous aging kinetics resulting from an approach based on a single time material is unable to capture the mentioned complex phenomenology.

Apart from the kinetics of equilibration under isothermal conditions, there exists at present increasing evidence that a single (α) relaxation process is unable to account for the phenomenology of physical aging. This evidence encompasses straightforward observation of a sub- T_g endotherm (also referred to as shadow glass transition) by calorimetry in glasses after aging well below T_g , as well as the increasing evidence that other mechanisms beyond the α relaxation mediate aging when this is explored over a wealth of experimental conditions.

Regarding the former aspect, the sub- T_g endotherm is a widely accepted feature observed in a variety of glasses, including bulk^{40–45} and confined⁸ polymers, metallic glasses,^{46–53} phase change materials,⁵⁴ chalcogenides,⁵⁵ and water based systems.⁵⁶ Furthermore, any attempts to catch this feature within a material time based approach required the use of *ad hoc* fitting parameters with questionable physical meaning; for instance, a non-exponentiality parameter as low as 0.2, providing unphysical stretching to the response.⁵⁷

The application of a single material time to describe aging in a wide range of conditions has been questioned for a long time.^{58,59} In recent times, a wealth of activity has therefore focused on the role of other mechanisms involved in the kinetics of glass equilibration. Depending on the time scale and the activation energy, this may range from the classical Johari–Goldstein (JG)⁶⁰ secondary relaxation,^{48,53,55,61–67} liquid-like features persisting deep in the glassy state,⁶⁸ to the recently identified slow Arrhenius process (SAP),^{69–73} bearing great potential in the equilibration of different kinetic processes.⁷⁴ Sharing the common feature of being localized motions,^{75,76} guidelines to identify the molecular mechanisms involved in the equilibration of amorphous systems have recently been provided.⁷⁷

VI. CONCLUSIONS

We have tested the ability of the SPA model with a single time scale to predict physical aging of small molecule glasses interacting via van der Waals forces. This was performed on a wide set of aging experiments, including previously published enthalpy relaxation data after temperature down-jumps and new results on enthalpy relaxation after temperature up-jumps. The general outcome of our study is that a single parameter is adequate to only partially capture the phenomenology of aging in glasses. Specifically, the single parameter able to describe aging after small temperature down- and up-jumps to the same aging temperature can capture aging

kinetics for relatively small temperature down-jumps but not for large ones. Conversely, it is possible to fit aging data over a wide range of temperature down-jumps, but the single parameter obtained from such a fit falls short of describing aging in temperature up-jump experiments.

The overall picture that can be drawn from our study is that the material time formalism with a single time scale accurately describes the phenomenology of aging for small jumps in proximity to the glass T_g .^{15–17,19} However, when aging is explored over a wider range of temperatures in both down-²² and up-jump^{78,79} experiments, the material time formalism requires refinements that account for the role of different time scales beyond that of the main α relaxation. Hence, we emphasize the need to include different “inner clocks” to account for different relaxation mechanisms involved in aging.⁸⁰

SUPPLEMENTARY MATERIAL

See the [supplementary material](#), which contains Fig. S1, which plots the fictive temperature evolution as a function of logarithmic aging time. It compares up-jump and down-jump aging experiments performed at the same aging temperature (T_a). The data are shown for all investigated systems, illustrating how fictive temperature changes with aging time in each case.

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AUTHOR DECLARATIONS

Conflict of Interest

The authors have no conflicts to disclose.

Author Contributions

All authors contributed equally to the study.

Valerio Di Lisio: Conceptualization (equal); Data curation (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Project administration (equal); Resources (equal); Software (equal); Supervision (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal). **Daniele Cangialosi:** Conceptualization (equal); Data curation (equal); Formal analysis (equal); Funding acquisition (equal); Investigation (equal); Methodology (equal); Project administration (equal); Resources (equal); Software (equal); Supervision (equal); Validation (equal); Visualization (equal); Writing – original draft (equal); Writing – review & editing (equal).

DATA AVAILABILITY

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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