Glassy dynamics in thin films of polystyrene

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Glassy dynamics was investigated for thin films of atactic polystyrene by complex electric capacitance measurements using dielectric relaxation spectroscopy. During the isothermal aging process the real part of the electric capacitance increased with time, whereas the imaginary part decreased with time. It follows that the aging time dependences of real and imaginary parts of the electric capacitance were primarily associated with change in volume (film thickness) and dielectric permittivity, respectively. Further, dielectric permittivity showed memory and rejuvenation effects in a similar manner to those observed for poly(methyl methacrylate) thin films. On the other hand, volume did not show a strong rejuvenation effect.

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I. INTRODUCTION

Amorphous materials show a glass transition as the temperature decreases in the liquid state. Throughout the glass transition, the molecular mobility due to the α-process is frozen and the material transforms into a glassy state. Polymeric glass shows structural changes during the aging process, even below the glass transition temperature $T_g$. Corresponding physical changes are observed with these structural changes [1, 2, 3]. These changes are known as physical aging, and are related to the glassy dynamics. This is regarded as an important property characteristic of disordered materials including: polymer glasses [4, 5, 6], spin glasses [7, 8] and other disordered systems [9, 10, 11].

In our previous papers [3, 4], the glassy states of poly(methyl methacrylate) (PMMA) show a decrease in both the real and imaginary parts of the complex dielectric constant with increasing time during the isothermal aging process. The glassy states show memory and rejuvenation phenomena for thermal treatment of a constant rate mode and of a temperature cycling mode, as shown below. In the constant rate mode, the temperature changes from high temperature above $T_g$ to an aging temperature, at which the sample is aged isothermally and the dielectric susceptibility decreases with increasing aging time during the process. This decrease in dielectric susceptibility is associated with the relaxation of the system into the equilibrium states. The change to the equilibrium is called aging. Then, the temperature subsequently decreases from the isothermal aging temperature to room temperature at a constant rate. A comparison of the value of the dielectric susceptibility at room temperature after the isothermal aging to that without isothermal aging, revealed that the values were in good agreement. This suggests that the decrease in dielectric susceptibilities induced by the isothermal aging is totally compensated during the cooling process from the isothermal aging temperature to room temperature. This change in dielectric susceptibilities has the opposite direction compared to the change due to the aging. Therefore, the change in dielectric susceptibility induced during the cooling process after the isothermal aging is called rejuvenation, and hence the system is rejuvenated as far as the dielectric response is concerned. However, during the subsequent heating process from room temperature, the dielectric susceptibility deviates from the value observed during the cooling process without isothermal aging, and shows a maximum deviation just above the aging temperature as if the sample remembered the isothermal aging it experienced during the preceding cooling process. This behavior is called memory effect and it suggests that the thermal history is maintained as a memory.

In the case of amorphous polymers, it is well known that the density of the polymers increases due to the isothermal aging below $T_g$, and this density change is maintained even at room temperature [1]. On the other hand, there is no rejuvenation for the density. This finding appears to be inconsistent with the fact that the dielectric as-susceptibility shows rejuvenation. Herein, we investigate the aging behavior of thin films of atactic polystyrene (a-PS) using electric capacitance measurements in order to elucidate the nature of the glassy dynamics in polymer glasses.

II. EXPERIMENTAL

Thin films of atactic polystyrene (a-PS) with two different thicknesses, 14 nm and 284 nm, were prepared by spin-coating from a toluene solution of a-PS on an Aluminum (Al)-deposited glass substrate. The thickness was controlled by changing the concentration of solution. The
samples of a-PS used in this study were purchased from Aldrich Co., Ltd. \(M_w = 1.8 \times 10^6, M_w/M_n = 1.03\). After annealing at 70 °C under vacuum for two days to remove solvents, Al was again vacuum-deposited to serve as an upper electrode. Several heating cycles through and above a bulk value of \(T_g\) were carried out before the capacitance measurements were taken for the relaxed as-spun films in order to obtain reproducible results. The values of \(T_g\) in the thin films with \(d = 14\) nm and 284 nm were 350 K and 368 K, respectively. The thickness was evaluated from the value of the electric capacitance at 293 K for the as prepared films before measurements in the same manner previously reported \[12,13\].

The \(T_g\) of the two thin films differs by about 18 K. According to the recent report on \(T_g\) of thin films of polystyrene, the effect of oxidation can be a possible origin of the reduction of \(T_g\) \[14\]. However, we performed \(T_g\) measurements for each thin film several times and confirmed that the reduction of \(T_g\) is not due to the oxidation effect in the present case.

Capacitance measurements were carried out using an LCR meter (HP4284A) for the frequency \(f = 1\) kHz during the cooling and heating processes between 380 K and 273 K at a rate of 1 K/min as well as during isothermal aging at various aging temperatures \(T_a\) \((=315 \text{ K} \sim 351 \text{ K})\). The complex electric capacitance of the sample condenser \(C^* (\equiv C' - iC'')\) was measured as a function of temperature \(T\) and aging time \(t_a\). The value of \(C^*\) was converted into the dynamic (complex) dielectric constant \(\epsilon^* (\equiv \epsilon' - i\epsilon'')\) by dividing the \(C^* (T)\) by the geometrical capacitance \(C_0(T_0)\) at a standard temperature \(T_0\). The value of \(C^*\) is given by the relation \(C^* = \epsilon^* \epsilon_0 \frac{S}{d}\) and \(C_0 = \epsilon_0 \frac{S}{d}\), where \(\epsilon_0\) is the permittivity in vacuum, \(S\) is the area of the electrode and \(d\) is film thickness. For evaluation of \(\epsilon^*\) and \(C_0\), we use the thickness \(d\) which is determined at \(T_0 = 293\) K and \(S = 8 \times 10^{-9} \text{ m}^2\).

**III. RESULTS AND DISCUSSION**

**A. Aging dynamics**

Figure 1 shows the change in the real and imaginary parts of the complex electric capacitance for a-PS films with \(d = 284\) nm and \(f = 1\) kHz. The temperature of the sample was changed as follows: After several temperature cycles in order to stabilize the measurements, the temperature changed from 380 K to 273 K (cooling process, dotted line) and then from 273 K to 380 K (heating process, solid line). The data obtained for the cooling and heating processes are used as a reference data. The sample was cooled from 380 K to 340 K(\(\equiv T_a\)), the temperature was maintained at \(T_a\) for 20 hours, and then the sample was again cooled down to 273 K after isothermal aging (open circles). Finally, the sample was again heated from 273 K to 380 K (open triangles).

In Fig. 1(a), the reference data shows that the real part of the electric capacitance \(C'\) is a decreasing function of temperature and the slope of \(C'\) with respect to temperature gradually changed around 365 K. Thus, there is a difference between the heating and cooling processes. This gradual change corresponds to the glass transition. Furthermore, from the data for the subsequent cooling process including isothermal aging, \(C'\) was observed to increase with aging time during isothermal aging at \(T_a\). In this case, the value of \(C'\) increased by 0.3 % for isothermal aging at 340 K for 20 hours.

Contrary to \(C'\), the imaginary part of the electric capacitance \(C''\) was an increasing function of temperature during the cooling and heating processes at a constant rate. Above 380 K a decrease in the peak was expected due to the \(\alpha\)-process. Further, during the isothermal aging process at 340 K, \(C''\) was observed to decrease with increasing aging time. The change in \(C''\) for aging at 340 K for 20 hours was about 0.2 pF, which corresponds to a decrease by 3%. This fraction was an order of magnitude larger than the increase in \(C'\) for the same aging process.

Figure 2 shows the temperature dependence of the complex electric capacitance in thin films of a-PS with \(d = 14\) nm. In this case, a similar result was obtained for the temperature dependence of \(C'\) and \(C''\) as that for \(d = 284\) nm. For thin films, the absolute value of \(C^*\) is enhanced. The change in \(C'\) is more distinct due to the smaller slope of \(C'\) with respect to \(T\). Furthermore, the scatter of data in \(C'\) is reduced due to the larger geometrical capacitance. The slight increase in \(C'\) above 375 K was due to the existence of the \(\alpha\)-process. In the case of
creases with aging time. It is important to note that both
served commonly for other polymeric systems. The observed in-
complex electric capacitances (dielectric constants) de-
it is also reported in the literature that the real part of
isothermal aging in poly(methyl methacrylate) \[4, 12, 13\].
of a-PS, 0.1 % and 3 % for the isothermal aging at 340 K for 20
hours, respectively.

This suggests that during the isothermal aging process
of a-PS, $C'$ increases with aging time, whereas $C''$ decreases
with aging time. It is important to note that both
$C'$ and $C''$ decrease with increasing aging time
for the isothermal aging in poly(methyl methacrylate) \[4, 12, 13\].

It is also reported in the literature that the real part
of complex electric capacitances (dielectric constants)
decreases with increasing aging time for polycarbonate \[15\]
and poly(ethylene terephthalate) \[10\]. The observed
increase in $C'$ in a-PS is different from the decrease
observed commonly for other polymeric systems.

Figure 3 shows the aging time dependence of $C'$ and
$C''$ during isothermal aging at various temperatures $T_a$
(between 321.3 K and 350.8 K) for $d = 14$ nm. The values
$\Delta C'(t_w)$ and $\Delta C''(t_w)$ are defined by $\Delta C'(t_w)=C'(t_w)-C'(0)$
and $\Delta C''(t_w)=C''(t_w)-C''(0)$, and the time $t_w=0$
is the initial time at which the temperature of the sample
reaches the aging temperature. In Fig. 3(a), the value
$\Delta C'$ increases monotonically with aging time and
the value $\Delta C'$ at 20 hours decreases with decreasing
aging temperature. In Fig. 3(b), it is found that $\Delta C''$ decreases
with increasing aging time and the absolute value of $\Delta C''$
at 20 hours decreases with decreasing $T_a$ between 341.2 K
and 321.3 K. The relaxation behavior of $C''$ for isother-
mal aging at $T_a=350.8$ K appears to be slightly different
from that at other $T_a's$ lower than 350.8 K.

B. Change in volume and dielectric permittivity

A possible explanation for the observed dependence
of $C'$ and $C''$ on the aging time during isothermal aging
is provided in this section. The real and imaginary parts
of the complex electric capacitance are given as follows:

\[
C'(\omega, T, t) = (\epsilon_\infty(T, t) + \epsilon_\text{disp}(\omega, T, t))C_0(T, t) \quad (1)
\]

\[
C''(\omega, T, t) = \epsilon''_\text{disp}(\omega, T, t)C_0(T, t), \quad (2)
\]

where $\epsilon_\infty$ is the dielectric constant at the high frequency
limit, and $\epsilon_\text{disp}$ and $\epsilon''_\text{disp}$ are frequency-dependent contribu-
tions to the dielectric constant due to the orientational
polarization associated with molecular motions. The fol-
lowing relations can be derived: $\epsilon' = \epsilon_\text{disp} + \epsilon_\infty$ and
$\epsilon'' = \epsilon''_\text{disp}$. In the case of a-PS, the polarity is very weak,
therefore; $\epsilon_\text{disp} \ll \epsilon_\infty$ is expected. Thus, Eq. (1) can be
approximately rewritten as

\[
C'(\omega, T, t) \approx \epsilon_\infty(T, t)C_0(T, t). \quad (3)
\]

For an isothermal aging process, the density is expected
to increase with aging time, and hence the film thickness
$d$ decreases on the condition that the area of the film
remains constant. Therefore, this density change causes
an increase in $C'$ according to the following rate

\[
\frac{1}{C'} \frac{\partial C'}{\partial t} = -(\eta + 1) \frac{1}{d} \frac{\partial d}{\partial t}, \quad (4)
\]
where \( \eta \) is a constant and its value is almost equal to 1 for a-PS \(^{12} \). Eq. (4) suggests that the decrease in film thickness increases \( C' \) for isothermal aging.

Figure 4 shows the temperature change in the inverse of \( C' \) observed during the constant rate mode. The vertical axis approximately corresponds to the volume of this system if the area remains constant. Therefore, in Fig. 4 a temperature dependence of the volume characteristic of amorphous polymers can be observed. As the sample is cooled down from a high temperature to a lower one through \( T_g \), the initial liquid state changes to a glassy state (Glass 1) and then the glassy state is converted into a second glassy state (Glass 2) during an isothermal aging. This temperature dependence of volume can typically be measured using dilatometric measurements for amorphous polymers.

On the other hand, Eq. (2) shows that \( C'' \) includes two different contributions from \( \epsilon''_{\text{disp}} \) and \( C_0 \). For the isothermal aging, \( C_0 \) is expected to increase with aging time, because the density increases, \( i.e., \) the film thickness decreases, as shown above. Therefore, the decrease in \( C'' \) with aging time suggests that the imaginary part of dielectric constant, \( \epsilon''_{\text{disp}} \), decreases with aging time and that its contribution can overcome the contribution from the increase in \( C_0 \). For PMMA, which has a strong polar group in the chain, it has been reported that both real and imaginary parts of the dielectric constant decrease with aging time during the isothermal aging process \(^{4, 6} \).

The results observed in a-PS can be explained as follows: for the isothermal aging process, the decrease in film thickness is observed as the increase in \( C' \), whereas the decrease in the imaginary component of dielectric permittivity is observed as a decrease in \( C'' \). Therefore, the present measurement on the electric capacitance of a-PS will provide information on the change in volume and di-lectric permittivity simultaneously for the same sample during the isothermal aging process.

### C. Volume relaxation during isothermal aging

As discussed in the previous section, the real part of the complex electric capacitance changes with aging time in accordance with the volume change. Figure 5 shows the aging time dependence of \( C' \) relative to the value of \( C'(0) \) for thin films with \( d = 284 \text{ nm} \). The horizontal axis is the logarithm of aging time. Figure 5 clearly shows that \( \Delta C' \) increases with the logarithmic law with respect to the aging time for the long time region. Further, there is no tendency to saturate in this time region. We evaluate the slope of this aging time dependence using the following equation:

\[
\Delta C'(t_w) = \hat{A} \log t_w + B \quad \text{(for large } t_w) \tag{5}
\]

Here, from the value of \( \hat{A} \), the aging rate of the volume, \( A \), for thin films of a-PS can be evaluated as follows:

\[
A = \frac{\hat{A}}{C'(0)} = \frac{1}{C'(0)} \frac{d\Delta C'}{d\log t_w} \tag{6}
\]

where for a given frequency \( C'(0) \) is the value of \( C' \) at the time at which the temperature reaches the aging temperature. Figure 6 shows the temperature dependence of \( A \) for thin films with two different thicknesses \( d = 14 \text{ nm} \) and \( 284 \text{ nm} \). In the present temperature range, the aging rate \( A \) was found to be larger in \( d = 284 \text{ nm} \) than in \( d = 14 \text{ nm} \). In this figure, it is clear that the rate \( A \) increases with increasing aging temperature and obeys the Arrhenius law: \( A = A_0 \exp(-U/k_BT) \), where \( k_B \) is the Boltzmann constant, \( U \) is the activation energy and \( A_0 \) is a constant.

It should be discussed what is the molecular origin associated with the volume change during the aging process. Below \( T_g \) there should be no contribution due to
the α-process, and hence it can be expected that the β-process should be the most probable candidate for the molecular origin for the physical aging. However, in the literature the activation energy for the β-process in atactic polystyrene was estimated to be 38 kcal/mol [17] or 30 kcal/mol [18], which are larger than the activation energy evaluated from the temperature dependence of the aging rate A. Therefore, the present result cannot be regarded as an evidence that the β-process is directly associated to the physical aging, although the α-process can be excluded from the candidate for molecular origin for the physical aging. Probably, the β-process is indirectly related to the volume change during the physical aging process.

D. Memory and rejuvenation in thin films

Figure 7 shows the temperature dependence of ∆C′ and ∆C″′ observed during the cooling process with isothermal aging at $T_a$ and the subsequent heating process for $f = 1$ kHz in α-PS films with $d = 284$ nm ((a), (b)) and 14 nm ((c), (d)). In this case, $\Delta C''(T)$ ($\Delta C'''(T)$) is evaluated as the deviation of $C'(T)$ ($C''(T)$) from the reference values $C'_{\text{ref}}(T)$ ($C''_{\text{ref}}(T)$) at the same temperature $T$. For the reference value $C'_{\text{ref}} (C''_{\text{ref}})$ to the cooling process including isothermal aging, we used the data measured for the preceding cooling process without any isothermal aging, and for the reference value for the heating process after the isothermal aging, we used the data measured for the preceding heating process.

Figure 7(a) shows that as the temperature decreases from 380 K to 340 K, $\Delta C'$ remains almost zero, and the deviation $\Delta C'$ increases as the aging time increases during isothermal aging at 340 K (See open circles). During the cooling process, after the isothermal aging, the deviation decreases and then approaches a constant value, but not zero. As a result, most of the deviation $\Delta C'$ induced during the isothermal aging remains even at 273 K. This result is thought to be associated with the fact that the isothermal aging increases the density. For the subsequent heating process, $\Delta C'$ changes along the path traced by $\Delta C'$ for the preceding cooling process after isothermal aging, and then $\Delta C'$ reaches a maximum just above $T_a$ (See open triangles). The value of $\Delta C'$ subsequently decreases approaching zero. This behavior can be interpreted as follows: the fact that the sample experiences aging at $T_a$ by the way of the preceding cooling process is memorized within the sample, and the memory is recalled during the subsequent heating process.

The temperature dependence of $\Delta C'''$ is different from that of $\Delta C'$, as shown in Fig. 7(b). For the cooling process from 380 K, $\Delta C'''$ remains almost zero, and then $\Delta C'''$ decreases with increasing aging time for the isothermal aging. $\Delta C'''$ subsequently increases with decreasing temperature and reaches zero at 273 K. In this case, there is a large scatter in the values of $C'$, because the thickness is 284 nm and the geometrical capacitance $C_0$ is not large enough. Figure 7(d) shows the temperature dependence of $\Delta C'''$ during the constant rate mode for $d = 14$ nm. For $d = 14$ nm, the scatter of $C'''$ is suppressed, therefore; it is clear that the value of $\Delta C'''$ reaches zero at 273 K after cooling from $T_a$. This result suggests that the system is rejuvenated as far as the dielectric response is concerned. For the subsequent heating process, $\Delta C'''$ decreases along the curve observed during the preceding cooling process, and reaches zero after showing a minimum just above $T_a$. Because the temperature dependence of $\Delta C'''$ is primarily attributed to that of $\varepsilon''$, it follows that $\varepsilon''$ exhibits a memory and rejuvenation effect. A similar effect has been observed for $\varepsilon'$ and $\varepsilon'''$ in the case of PMMA [1, 6].

Combining the results observed for $\Delta C'$ and $\Delta C'''$, it is concluded that the volume of α-PS films decrease during isothermal aging, and the deviation from the reference value is maintained below the aging temperature. On the other hand, dielectric permittivity also decreases during isothermal aging, and the deviation of the dielectric permittivity from the reference value is totally rejuvenated at lower temperatures. The existence of the volume change observed in the present measurement is consistent with the fact that there are several reports relating to the change in volume or density due to physical aging [1]. Based on the volume, the system does not appear to be rejuvenated, however, the dielectric response to the ac-electric field is fully rejuvenated.

In disordered systems, similar memory and rejuvenation effects are often observed during the aging process. In spin glasses, magnetic ac-susceptibility shows a memory and rejuvenation effect during the constant rate mode and the temperature cycling mode. A possible mechanism for this effect has been extensively discussed by a number of research groups [19, 20, 21, 22, 23, 24, 25].

In spin glasses, memory and rejuvenation effects have been investigated through the measurements of magnetic ac-susceptibility. A number of experiments clearly show the existence of a strong rejuvenation effect in aging phenomena through the so-called temperature-cycling mode, where the aging temperature is maintained at $T_1$ for the
FIG. 7: Temperature dependence of the deviation $\Delta C'$ ($\Delta C''$) of the component of the complex electric capacitance $C'$ ($C''$) from the reference values observed for the cooling process including at an isothermal aging at 350.8 K and the subsequent heating process for thin films of a-PS with $d = 14$ nm and $f = 1$ kHz ((a) and (b)) and 100 Hz ((c) and (d)).

First and third stage and at $T_2$ (smaller than $T_1$) for the second stage. At the beginning of the second stage of the temperature-cycling mode, the imaginary part of the magnetic ac-susceptibility $\chi_{ac}$ returns to the initial value for $\chi_{ac}$ at the beginning of the first stage, although $\chi_{ac}$ decreases during the first stage. However, it should be noted here that the magnetic ac-susceptibility is associated with a time scale, defined by $t_\omega = 2\pi/\omega$ (where $\omega$ is the angular frequency of the ac-magnetic field). There is a relationship between the time scale and the length scale $\xi(t_\omega)$, therefore, the magnetic ac-susceptibility is associated with the length scale $\xi(t_\omega)$. The experimental results suggest that the dielectric ac-susceptibility shows a rejuvenation effect, whereas the volume of the system changes with increasing aging time and shows no rejuvenation. Although these findings appear to be inconsistent, this can be explained as follows. Similar to magnetic ac-susceptibility, the dielectric ac-susceptibility is associated with a length scale $\xi(t_\omega)$ defined by the angular frequency $\omega$ of the applied electric field. The volume or density is a macroscopic physical quantity and has a characteristic length scale significantly larger than $\xi(t_\omega)$ for the present frequency range. This suggests that, even if the volume changes during the isothermal aging process and the change is maintained at lower temperatures, the dielectric response to the applied electric field is not influenced by the change in volume. Therefore, the dielectric ac-susceptibility can be rejuvenated during the cooling process after the isothermal aging process.

If both magnetic ac-susceptibility and a macroscopic physical quantity such as the remaining magnetization are measured simultaneously during the isothermal aging process, it can be expected that for spin glasses the magnetic ac-susceptibility will show a rejuvenation, whereas the magnetization is not rejuvenated.

IV. SUMMARY

Herein, we investigated the aging dynamics for thin films of atactic polystyrene through measurements of complex electric capacitance using dielectric relaxation spectroscopy. The results obtained in this study are summarized as follows:

1. During the isothermal aging process at a given aging temperature the real part of the electric capacitance increases with aging time, while the imaginary part decreases with aging time.

2. The aging time dependences of the real and imaginary parts of the electric capacitance are mainly associated with the change in volume or film thickness and dielectric permittivity, respectively.

3. Memory effect can be observed for both ac-dielectric permittivity and volume. On the other hand, a strong rejuvenation effect can be observed for the ac-dielectric permittivity, but not for the volume.
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