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Azo Polymers for Reversible Optical Storage. 9. Copolymers Containing Two Types of Azobenzene Side Groups

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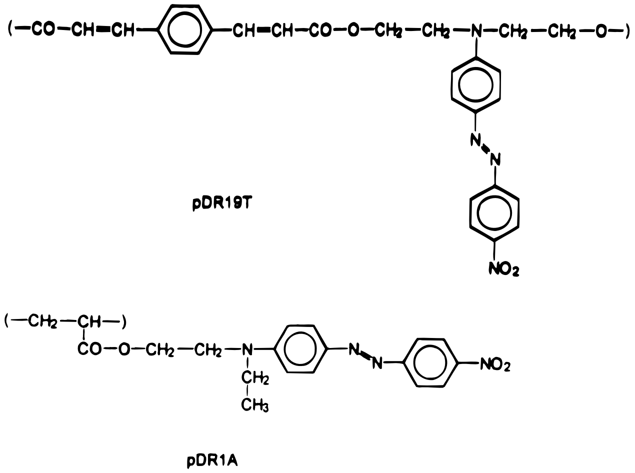
ABSTRACT: A series of copolymers with two structural units—1-(4-nitrophenyl)-2-(4-{[2-(methacryloyloxy)-ethyl]ethylamino}phenyl)diazene (DR1M), which contains donor−acceptor substituents in the azobenzene group, and 4-[2-(methacryloyloxy)ethyl]azobenzene (MEA), which has no donor−acceptor substituents—was prepared. The maximum obtainable photoinduced birefringence and the fraction of birefringence conserved after relaxation increase with increasing DR1M concentration in the copolymer. The rate of inducing birefringence is independent on the copolymer composition. In terms of the relaxation of the photoinduced birefringence, copolymers with higher DR1M content relax faster. The β-parameter of the stretched exponential functions describing the growth and relaxation of the photoinduced birefringence also appears to be independent of copolymer composition. The electronic spectra, as well as the birefringence studies, reveal that there is no significant interaction between DR1M and MEA structural units in the copolymers.

Introduction

Azo polymers are receiving much attention because of their potential uses in various photonic applications.[1,2] In our laboratory, amorphous high-T_g azo polymers are investigated as materials for reversible optical storage, mainly by studying the stable photoinduced birefringence and the structural parameters affecting the process of inducing and erasing the birefringence.[3−8] Birefringence is induced by a series of photochemically activated trans−cis−trans isomerization cycles accompanied by a slight motion of the azobenzene groups. Using polarized light, only azobenzene groups with dipoles having components along the laser polarization will be activated for isomerization. At the end of each trans−cis−trans isomerization cycle, an azobenzene group which has fallen perpendicular to the laser polarization will become inactive, and the concentration of the inactive groups will grow while the laser is on. The result is an anisotropy induced in the orientation of the azobenzene groups, which could be measured either as dichroism or as birefringence (the oriented azobenzene groups change the refractive index of the film). A significant fraction of this anisotropy (typically 80% of the birefringence) is stable for an infinitely long time at room temperature, if the glass transition temperature of the polymer is relatively high (at least 80−90 °C). On this "written" film, the same laser with circularly polarized light will activate all azobenzene groups for isomerization and will restore the original random orientation, thus "erasing" the birefringence. The phenomenon is perfectly reversible and the same spot on the film can be written and erased for tens of thousands of times.

There are a series of structural and physical factors which affect the level and rate of growth of birefringence, as well as the long-term stability of the induced supramolecular organization. Several such factors have been identified in our laboratory. The nature of the azo group is very important, because it determines the laser wavelength that can be used for photoinducing birefringence. Most of the azo groups used have donor−acceptor substituents,[4,5] but no such substituents are necessary,[8] although if they are absent, the process is much slower and not necessarily reproducible and the levels of birefringence are lower. The film thickness is very important,[6] since the azo chromophores absorb the laser light, thus creating a nonlinear distribution of light intensity within the film. The azo content is very important, with more azo groups within the polymer producing higher levels of birefringence.[9] The glass transition temperature of the polymer, above which thermal motion randomizes any induced orientation, is obviously a very important structural factor dictating the long-term stability of the induced birefringence.

Chart 1


pDR19T

pDR1A

Two structural factors are receiving special attention, since they allow control over the levels and rates of induced birefringence. The first factor is the thermodynamic tendency of the polymer to form organized domains. If the polymer is crystalline or liquid crystalline, the photoinduced supramolecular organization is reinforced and the levels of birefringence attainable in such systems are significantly higher than in amorphous polymer films. For example, the amorphous pDR1A (Chart 1) gives a maximum birefringence of about 0.08, while pDR19T,[7] which is about 16% crystalline, produces a birefringence level of about 0.27. These levels are highly stable for long times, and the disadvantage of the crystalline/liquid crystalline materials is

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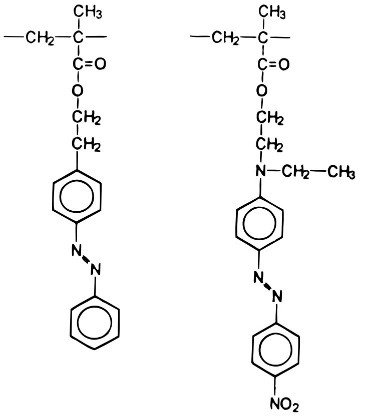
Chart 2



MEA DR1M

that it takes much longer to induce the birefringence, since the process involves complex motions in order to form whole crystalline (liquid crystalline) domains. Also, it is difficult and time-consuming to “erase”, since randomness is never restored; only the directionality of various crystalline domains is randomized.

The second factor is the existence of cooperative motions of neighboring groups, which may enhance the orientation. This factor is well known in crystalline and liquid crystalline materials and—to some extent—is similar to the above description. In liquid crystalline polymer films containing azo and ester/amide mesogens,[10] changing the direction of the azo mesogens using polarized light will activate the photoinactive mesogens to move in concert, thus re-creating the initial liquid crystalline order. A similar phenomenon has been described in azo-containing Langmuir–Blodgett films,[11] where liquid crystals can change their orientation by sympathy with the trans–cis isomerization of the adjacent azobenzenes. Recently, we have observed that such a cooperative motion exists in amorphous polymers as well. In amorphous copolymers of 1-(4-nitrophenyl)-2-(4-{[2-(methacryloyloxy)ethyl]ethylamino}-phenyl)diazene (DR1M) with methyl methacrylate (MMA), neighboring azo groups tend to move in concert, while isolated azo groups are much easier to move.[9] There are two possible explanations for this phenomenon, since these polymers have no intrinsic order of the crystalline or liquid crystalline type. The groups may be influenced by the electric field of the neighboring dipole or there may be a steric effect requiring them to move together. The only way to distinguish between these two possible factors is to synthesize various copolymer pairs, trying to eliminate either the dipolar or the steric factor.

This paper presents the synthesis and photoinduced birefringence of a series of copolymers of DR1M with 4-[2-(methacryloyloxy)ethyl]azobenzene (MEA). The formulas of the two structural units are presented in Chart 2. This system was chosen to test a combination of polar/less polar structural units of similar shapes, where the steric effects should be more important. In addition, MEA exhibits a very different response to the visible laser in comparison to DR1M, as shown by the homopolymer systems.[12]

Experimental Section

The monomers, 1-(4-nitrophenyl)-2-(4-{[2-(methacryloyloxy)-ethyl]ethylamino}phenyl)diazene (DR1M)[13] and 4-[2-(methacryloyloxy)ethyl]azobenzene (MEA),[8] were prepared as previously reported. The polymerizations were carried out in dry toluene (0.15 M) with 10 wt % AIBN as initiator. The monomers were allowed to copolymerize under argon at 60 °C for 2 days. The polymerization was stopped by pouring the reaction mixture into methanol. The copolymers were redissolved in THF and precipitated again in methanol, and finally dried under vacuum.

Table 1. Weight-Average Molecular Weights (M_w), Polydispersity Indices (M_w/M_n), and Glass Transition Temperatures (T_g) of the Copolymers

polymer	M_w	M_w/M_n	T_g (°C)
pMEA	70 000	2.2	80
D1	20 000	2.3	80
D2	16 000	2.1	86
D3	18 000	2.3	91
D4	21 000	1.9	100
D5	11 000	1.7	99
D6	20 000	2.2	105
D7	22 000	2.6	109
D8	25 000	2.4	114
D9	19 000	2.0	119
pDR1M	9 000	1.8	116

The proton spectra were recorded on a Bruker AC-F 200 NMR spectrometer. Electronic spectra were obtained on a Hewlett-Packard 8452A diode array spectrophotometer. The average molecular weights (relative to polystyrene) were obtained on a Waters Associates liquid chromatograph equipped with μ-Styragel columns and an R401 differential refractometer. The glass transition temperatures (T_g) were obtained on a Mettler TA 3000 thermal analysis system equipped with a TC10A processor and a DSC3 head at a heating rate of 20 K/min in the second scan. The average molecular weights and the T_gs of the copolymers are summarized in Table 1. All copolymers are completely amorphous as determined by DSC and polarized microscopy.

Thin films of the copolymers were obtained by dissolving the copolymers in THF and spin-coating onto clean glass slides. The films were allowed to dry and subsequently were heated above the T_g of the copolymers. Relatively homogeneous thin films from 100 to 400 nm thickness were obtained by this procedure. Film thickness was determined by interferometry. The detailed procedure for the measurement of the optically induced birefringence[3-6] has been described previously.

Results and Discussion

Copolymer Characterization. The copolymer compositions are determined from ^{1}H-NMR spectra. As an example, the NMR spectrum of copolymer D4 is shown in Figure 1. The assignments are made by comparison with the spectra of the homopolymers. The compositions can be readily obtained using ratios of the integrated peak areas in the aromatic and aliphatic regions. The results are listed in Table 2. From the α-methyl signal, it appears that the tacticity of all these copolymers is similar to the tacticity of pMMA obtained by free-radical mechanisms, i.e., predominantly syndiotactic.

The reactivity ratios of the high-conversion copolymerization are estimated by the Kelen–Tudos high-conversion formula.[14] The values are $r_{MEA} = 0.47$ and $r_{DR1M} = 0.33$. This suggests that the copolymers have a random distribution with a trend toward alternating structural units.

The UV–vis spectra for selected samples of the copolymer series are shown in Figure 2. There is a shift of the absorption maximum in the visible region to shorter wavelengths as the DR1M content in the copolymer increases, by a maximum of 10 nm in comparison with pDR1M. The absorption maximum in the UV region does not seem to be affected by the composition. These observations suggest that there is

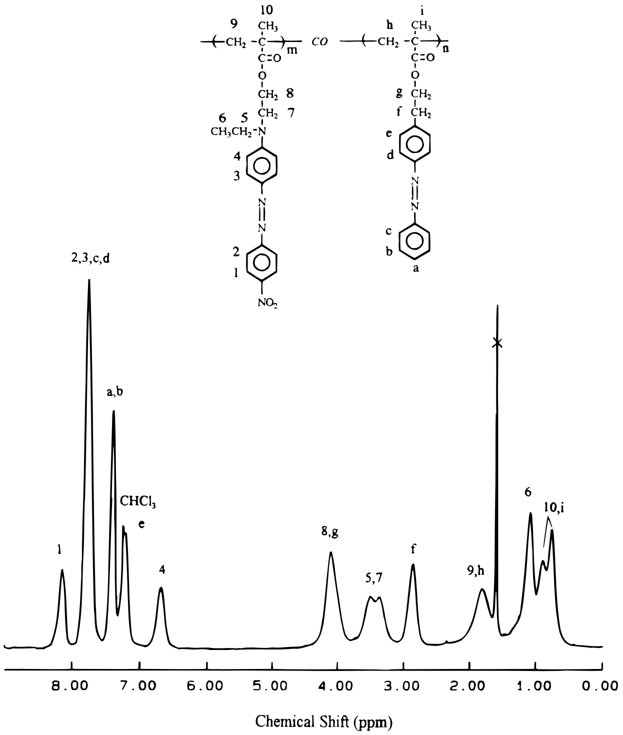


Figure 1. ^{1}H-NMR spectrum of poly(DR1M-*co*-MEA) sample D4.

Table 2. Details of Polymerization

copolymer	initial feed (MEA mol fract)	copolymer comp (MEA mol fract)	conv (wt fract)
D1	0.90	0.87	0.71
D2	0.80	0.78	0.64
D3	0.70	0.68	0.73
D4	0.60	0.57	0.81
D5	0.50	0.52	0.83
D6	0.40	0.42	0.76
D7	0.30	0.34	0.80
D8	0.20	0.24	0.82
D9	0.10	0.13	0.71

no significant interaction between these two kinds of azobenzene groups in the copolymers. This is unlike the copolymer series containing DR1M and MMA,[9] which exhibits a red shift of up to 22 nm in comparison to pDR1M. On the other hand, a red shift of only 10 nm was observed in the polymer blends containing pDR1A and pMMA. Another copolymer series, containing DR1M and a polar group, 4-nitrophenyl 4-[2-(methacryloyloxy)ethyl]benzoate, exhibits a red shift of up to 35 nm.[15]

Reversible Optical Storage. A writing laser beam with a wavelength of 514 nm (0.7 W/cm^2) was used to induce birefringence in all polymer films. Similar to the previously reported polymer systems, birefringence can be induced by a linearly polarized laser beam and the photoinduced birefringence can be “erased” by a circularly polarized beam. Figure 3 shows a typical sequence of inducing and erasing birefringence for copolymer D6. Birefringence is induced to a saturated level by a linearly polarized laser beam and relaxation occurs when the laser beam is turned off at 27 s. The circularly polarized laser beam is turned on at 96 s and the birefringence is erased.

Birefringence Levels. The maximum birefringence attained in the copolymer films and the birefringence conserved after relaxation are plotted as a function of DR1M content in Figure 4. Both increase monotonically with the DR1M content in the copolymer. Since the birefringence level obtained in pDR1M is 0.1, while on pMEA films in the same conditions the level is one order of magnitude lower, this dependence is not surprising. The explanation is related to the increase in the polarity of the side groups. It has been demonstrated that the level of birefringence attainable in a polymer film in the above-described conditions is higher for azobenzene side groups of higher polarity.[12] A simple explanation is the increased absorbance at the laser wavelength, which increases the rate of the trans–cis isomerization process, but dipolar associations of the side groups also favor increased birefringence levels.

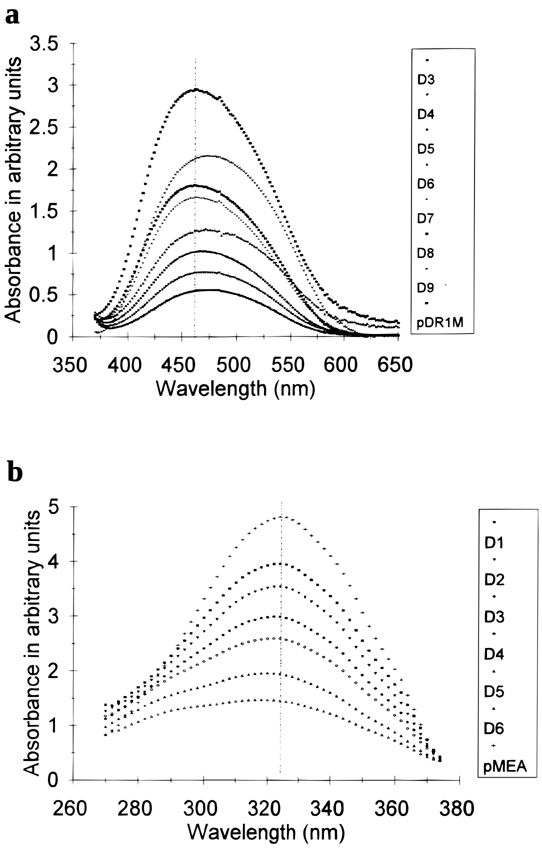


Figure 2. (a) Comparison of absorption maxima in the visible region for seven copolymer samples and the homopolymer pDR1M. The vertical dashed line indicates the wavelength of maximum absorbance of pDR1M. (b) Comparison of absorption maxima in the UV region for six copolymer samples and the homopolymer pMEA. The vertical dashed line indicates the wavelength of maximum absorbance of pMEA.

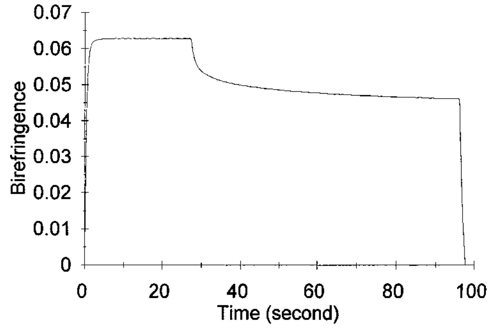


Figure 3. Write–erase profile for optically induced and erased birefringence of poly(DR1M-*co*-MEA) sample D6 with a laser beam at 514 nm. Time = 0 s: linearly polarized laser ON; time = 27 s: laser OFF; time = 96 s: circularly polarized laser ON.

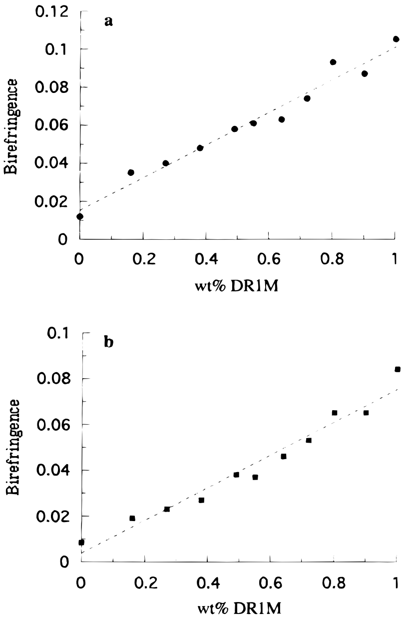


Figure 4. (a) Maximum induced birefringence values for the copolymer series plotted as a function of DR1M weight fraction. The line is drawn to guide the eye. (b) Birefringence conserved after relaxation for the copolymer series plotted as a function of DR1M weight fraction. The line is drawn to guide the eye.

The birefringence also increases linearly with the DR1M content in the copolymer. Such a linear dependence has also been observed for the polymer blends of pDR1A with pMMA, while the copolymers p(DR1A-*co*-MMA) and p(DR1M-*co*-MMA) show a nonlinear dependence, which has been explained by intramolecular interactions between the side groups.[9] The results presented in Figure 4 suggest that there is no observable cooperative effect in this copolymer series. In order to confirm this, blends of pDR1M and pMEA should be studied and compared with the copolymer series; unfortunately, these two homopolymers are not miscible and do not yield transparent films; thus no comparison data are available.

Growth of the Photoinduced Birefringence. It is important to know how fast the birefringence can be induced in the film by the polarized laser. We have recently reported that the growth of birefringence can be described by a biexponential function[12,13] consisting of a fast process mainly involving the motions of the azobenzene side group (and thus depending on the quantum yields of isomerization, isomerization rates, and the local free volume around the azo group) and of a slow process involving some motions of the main chain (and thus depending on the coupling interaction between the azo group and the main chain and on the segmental mobility of the polymer). This biexponential function works well for a series of homopolymers and even for copolymers[9,15] but has no physical meaning for the present copolymer series. Indeed, the two types of azobenzene present here have been shown to behave quite differently when irradiated at 514 nm. MEA groups absorb very little at 514 nm, with the consequence that the photoinduced trans−cis isomerization is less efficient and the birefringence is induced at a slower rate. Thus, two very different isomerization rates could not be approximated by a single fast process, and the results are scattered when a biexponential fit is attempted. The overall response of the copolymer to the laser beam is much more complicated than that of the homopolymers containing only one type of the photoresponsive group, and simple biexponential approximations are not sufficient to describe the birefringence growth. On the other hand, the Kohlrausch−Williams−Watts (the stretched exponential) function,[16] which characterizes the growth process as a distribution of time constants would give a more meaningful description of the process. Equation 1 was found to give a reasonably good fit to the growth of the photoinduced birefringence. Figure 5a shows one example of curve fitting.

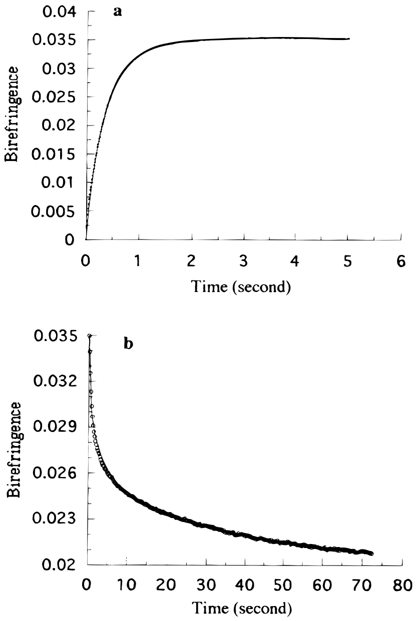


Figure 5. (a) Photoinduced birefringence growth curve for poly(DR1M-*co*-MEA) sample D1 (solid circles). The solid line is a fit to eq 1. (b) decay of the birefringence after the writing beam is turned off for poly(DR1M-*co*-MEA) sample D1 (open circles). The solid line is a fit to eq 2.

Table 3. Parameters Obtained by Fitting the Birefringence Growth Curves to Eq 1

polymer	A	τ_w (s)	β_w
pMEA	0.012	$0.806 \pm 8 \times 10^{-3}$	$0.89 \pm 1 \times 10^{-2}$
D1	0.035	$0.360 \pm 4 \times 10^{-4}$	$0.88 \pm 1 \times 10^{-3}$
D2	0.040	$0.350 \pm 3 \times 10^{-4}$	$0.89 \pm 1 \times 10^{-3}$
D3	0.048	$0.336 \pm 3 \times 10^{-4}$	$0.90 \pm 1 \times 10^{-3}$
D4	0.058	$0.317 \pm 2 \times 10^{-4}$	$0.90 \pm 1 \times 10^{-3}$
D5	0.061	$0.323 \pm 3 \times 10^{-4}$	$0.88 \pm 1 \times 10^{-3}$
D6	0.063	$0.314 \pm 2 \times 10^{-4}$	$0.89 \pm 1 \times 10^{-3}$
D7	0.074	$0.323 \pm 3 \times 10^{-4}$	$0.87 \pm 1 \times 10^{-3}$
D8	0.093	$0.323 \pm 2 \times 10^{-4}$	$0.91 \pm 1 \times 10^{-3}$
D9	0.087	$0.305 \pm 2 \times 10^{-4}$	$0.89 \pm 2 \times 10^{-3}$
pDR1M	0.103	$0.274 \pm 3 \times 10^{-3}$	$0.78 \pm 1 \times 10^{-2}$

$$y = A\{1 - \exp[-(t/\tau_w)^{\beta_w}]\} \tag{1}$$

In eq 1, y is the birefringence, A is a parameter representing the maximum achievable photoinduced birefringence, τ_w is the characteristic growth time, and β_w is the width of the distribution of the growth times. Table 3 summarizes the fitting parameters for the copolymer series. Although the maximum achievable birefringence increases monotonically with the DR1M content in the copolymer, the width of distribution of growth times (β_w) shows no dependence on the copolymer composition. This indicates that the processes involved in the optically induced birefringence are essentially the same throughout the copolymer series. In other words, there is no significant interaction between the two kinds of azobenzene groups in the

Table 4. Parameters Obtained by Fitting the Birefringence Decay Curves to Eq 2

polymer	B	C	τ_r (s)	β_r	C_n[a]
pMEA	0.004	0.008	7.9 ± 0.4	0.40 ± 0.01	0.67
D1	0.016	0.019	8.0 ± 0.4	0.37 ± 0.01	0.54
D2	0.017	0.023	6.5 ± 0.4	0.38 ± 0.01	0.58
D3	0.021	0.027	5.4 ± 0.2	0.40 ± 0.01	0.56
D4	0.020	0.038	5.9 ± 0.3	0.43 ± 0.01	0.66
D5	0.024	0.037	5.7 ± 0.2	0.39 ± 0.01	0.61
D6	0.017	0.046	5.8 ± 0.2	0.41 ± 0.01	0.73
D7	0.021	0.053	5.1 ± 0.2	0.47 ± 0.01	0.72
D8	0.028	0.065	5.1 ± 0.2	0.40 ± 0.01	0.70
D9	0.022	0.065	5.0 ± 0.2	0.41 ± 0.01	0.75
pDR1M	0.018	0.082	4.2 ± 0.3	0.41 ± 0.01	0.82

[a] $C_n = C/(B + C)$.

polymer matrix during the "writing" process. Although the characteristic growth time (τ_w) shows a slight decrease with increasing DR1M content in the copolymer, the change is not very significant. The "writing" process should also be related to the overall mobility of the polymer matrix. It is expected that the higher the molecular weight and the glass transition temperature (T_g) of a polymer, the more difficult such motions are. The copolymers have comparable molecular weights and the T_gs increase with the DR1M content. From the results obtained, however, the T_g does not seem to have a pronounced effect on the "writing" behavior in this copolymer series.

Relaxation of the Induced Birefringence. When the "writing" laser beam is turned off, the birefringence decays to a stable level. It is important to know how fast the birefringence decays (relaxes) and what level of birefringence will be stable for a long time. We have recently reported that the birefringence relaxation can also be described by a biexponential function[12,13] consisting of a fast process mainly involving the motions of the azobenzene side group (and thus depending on the thermal cis–trans isomerization rates and the local free volume around the azo group) and of a slow process involving some motions of the main chain (and thus depending on the coupling interaction between the azo group and the main chain and on the segmental mobility of the polymer). As for the birefringence growth, this biexponential function also works well to describe the birefringence relaxation of a series of homopolymers and even of some copolymers[9,15] but has not physical meaning for the present copolymer series. Indeed, the thermal cis–transition isomerization rates differ by orders of magnitude in DR1M (rate constant = 0.22 s^{-1}) and MEA (rate constant = 1.9×10^{-4} s^{-1}), with DR1M isomerizing much faster;[17] thus it would be impossible for these two processes to be described by a single fast rate in a biexponential description. Again, a stretched exponential must be used to describe the relaxation process:

$$y = B \exp[-(t/\tau_r)^{\beta_r}] + C \qquad (2)$$

where B represents the relaxation of the induced birefringence after the linearly polarized laser beam is off, τ_r is the characteristic relaxation time, and β_r is the width of the distribution of relaxation times. In addition, parameter C represents the induced birefringence conserved after relaxation. The fitting parameters for eq 2 are summarized in Table 4, while Figure 5b shows an example of a fitted relaxation curve.

As shown in Table 4, the characteristic relaxation time decreases as the content of DR1M in the copolymer increases. In other words, copolymers with higher DR1M content relax faster. Since thermal cis–trans isomerization is a very important component of relaxation, this is an expected trend based on thermal cis–trans isomerization constants in the two homopolymers.[17] In addition, the effect of molecular associations during dipole reorientation should also be considered. Copolymer with higher DR1M content should have stronger molecular associations, based on the larger DR1M dipole moment; thus both these factors might account for the differences in the characteristic relaxation times of the copolymer series. The overall mobility of the polymer matrix is also a factor influencing the relaxation process, and it would be affected by the molecular weight and by the T_g of the polymer. These copolymers have comparable molecular weights and their T_gs increase with the DR1M content. However, the copolymer with the lowest DR1M content, with the lowest Tg, has the longest τ_r. This result also suggests that T_g is not one of the most important factors in the relaxation behavior; probably the thermal cis–trans isomerization is the dominant factor.

The parameter C in eq 2 is time independent; thus it represents the birefringence conserved in the polymer film after relaxation. It is interesting to note that the fraction of induced birefringence conserved (C_n) after relaxation increases with the DR1M content in the copolymer (Table 4). This suggests that dipolar interactions are important to stabilize the local ordering. In addition to the dipolar interaction, the T_g of the copolymer also affects the stability of the local ordering, because the orientational ordering conserved will depend on the difference between the operating temperature and T_g, and copolymers with higher DR1M contents have higher T_gs.

The parameter β_r of the stretched exponential function gives values of about 0.4, which are in agreement with those found for relaxation processes in other azo-containing copolymers using forced Rayleigh scattering[18] and pyroelectrical measurements.[19]

Conclusions

The saturated levels of birefringence increase linearly with the DR1M content in the copolymers. In addition, the fraction of birefringence conserved after relaxation follows the same trend. The composition of the copolymer does not have a significant effect on the characteristic growth time of the photoinduced birefringence for the copolymer series studied. On the other hand, the characteristic relaxation time is lower (faster relaxation) for higher DR1M contents in the copolymer. Both widths of distribution of the birefringence growth and relaxation times (β_w and β_r) show no essential dependence on the composition of the copolymer. Results from the UV–vis spectra and birefringence studies suggest that there is no significant interaction between the DR1M group and the MEA group in the copolymers. This confirms previous results which indicate that the dipolar effect is more important than the steric effect in cooperative motions of side groups in azo-containing polymers.[9,15]

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