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Hyun Taeg Hong^a, Kang Jin Cho^a & Dong Hoon Choi^a

^a College of Environment and Applied Chemistry, Institute of Material Science and Technology, Kyung Hee University, Yongin-shi, Kyungki-Do, 449-701, KOREA

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Orientational Relaxation Dynamics in Methacrylate Polymers by Means of *In-Situ* Measurement of the Electro-Optic Property

HYUN TAEG HONG, KANG JIN CHO and DONG HOON CHOI

*College of Environment and Applied Chemistry, Institute of Material Science and
Technology, Kyung Hee University, Yongin-shi, Kyungki-Do 449-701, KOREA*

Side chain second-order nonlinear optical (NLO) polymers were synthesized for studying the relaxation dynamics of the electro-optic effect. 2-(Methyl-4-[2-(4-nitrophenyl)-vinyl]-phenyl)-amino)-ethanol was synthesized to be bound to methacrylate repeating unit. We synthesized homopolymer and copolymer to compare the temporal relaxation behavior of second-order NLO properties. For this purpose, a real time poled and probe technique was employed to investigate the relationship between the measurement temperature and the electro-optic (E/O) property. The relaxation behavior of the E/O effect was analyzed below and above the glass transition temperature.

Keywords: side chain polymer; temporal stability; electro-optic effect

INTRODUCTION

Organic second-order nonlinear optical (NLO) materials have recently been attracting attention and provide strong potential advantages for second harmonic generation (SHG) and electro-optic (E/O) applications.^[1-2] Particularly, poled polymeric systems have drawn

remarkable interests as promising candidates for application in electro-optic and photonic devices. 2-(Methyl-{4-[2-(4-nitrophenyl)-vinyl]-phenyl}-amino)-ethanol chromophore was utilized in the fabrication of polymeric materials with large, stable optical nonlinearity. In an attempt to investigate the relaxation dynamics of the E/O effect after poling, we designed and synthesized two polymers. One is a copolymer with methylmethacrylate and the other is a homopolymer. Using two polymers, we did *in-situ* measurement of the linear E/O effect and traced the relaxation behavior of E/O signal at the temperature higher and lower than the glass transition temperature.^[3,4]

EXPERIMENTAL

Two polymers were prepared by following the known method. The structures of polymers were illustrated in Figure 1. Films were prepared by spin coating from a concentrated solution of approximately 15% by weight of polymers. The thermal behavior of polymers were investigated

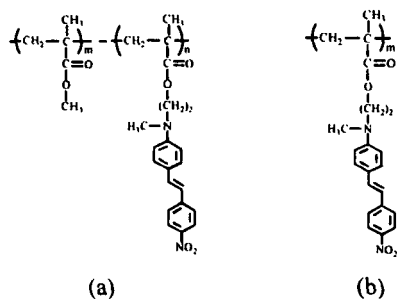


FIGURE 1 Structures of polymers used in this study; (a) copolymer m:n=2:1; (b) Homopolymer.

with differential scanning calorimetry. We investigated the relaxation dynamics of the E/O effect *via* a new measurement technique.^[3,4]

RESULTS AND DISCUSSION

Two polymers were utilized for investigating the change of the E/O effect with the temperature. Copolymer and homopolymer possess T_g at 110 and 107 °C respectively. In each relaxation measurement of E/O response, the sample temperature was raised upto 130-133°C that is higher than T_g with a continuous poling voltage applied between the electrodes. The sample was poled for 5 min, then the temperature is decreased down to the desired temperature, T while the poling voltage is maintained. The poling voltage is further maintained for an additional 5 min in order to let the sample equilibrate thermally and then switched off. At the same time, we applied the a.c. voltage of amplitude 10V and measured the decay of the E/O signal with respect to time.

In the cases of two polymers, we found that the isothermal relaxation of the E/O response can be described by KWW stretched exponential function with temperature dependent average decay time constant $\langle\tau\rangle$ and stretching parameter β . The relaxation of the E/O effect is shown to follow an Arrhenius temperature dependence below T_g , while it can be described by using a Vogel-Fulcher-Tamann-Hesse(VFTH) theory above T_g . First, below T_g , we investigated the relationship between the average relaxation time and the measurement temperature T . From the above relationship, activation energy, E_a of the copolymer and homopolymer were determined to be 62.9 and 67.4 kcal/mole respectively. From the results, the homopolymer possesses higher E_a for the local motion in the side chain. It is ascribed to the extent of dipolar interaction between the polar chromophores. Above T_g , we also could

fit the decaying data to the VFTH equation. The homopolymer showed larger decrement of the time constant with the temperature. It indicates that larger free volume change took place per unit temperature rise.

The temporal stability of NLO effect in the poled polymer is strongly dependent on the geometrical structure and the molecular interaction of the side chains and the main chains over a wide range of the temperature.

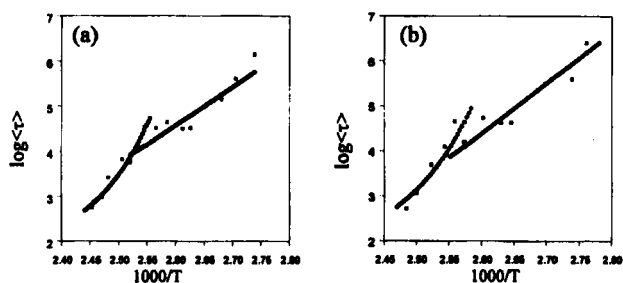


FIGURE 2 Dependence of the average time constant as a function of the inverse of the temperature T . (a) Copolymer; (b) Homopolymer.

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