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Publisher: Taylor & Francis

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Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals

Publication details, including instructions for authors and subscription information:

<http://www.tandfonline.com/loi/gmcl19>

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Version of record first published: 04 Oct 2006

To cite this article: Domasius Nwabunma, Zeng Long Zhou & Thein Kyu (1997): Development of Non-Birefringent Optical Adhesives, Molecular Crystals and Liquid Crystals Science and Technology. Section A. Molecular Crystals and Liquid Crystals, 301:1, 301-305

To link to this article: <http://dx.doi.org/10.1080/10587259708041781>

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DEVELOPMENT OF NON-BIREFRINGENT OPTICAL ADHESIVES

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Abstract The birefringence compensation technique¹ has been employed through polymer blending for the development of non-birefringent optical adhesives. It has been found that trimellitic anhydride(TMA)-cured polymethylmethacrylate(PMMA)/Epoxy (EPON-828) as a possible system resulted in phase separation due to polymerization². To circumvent the problem of polymerization-induced phase separation, a copolymer system comprised of methylmethacrylate(MMA) and glycidylmethacrylate(GMA) was synthesized³. The MMA-co-GMA copolymer was subsequently blended with EPON-828, then cured thermally with TMA. The cured blend films showed single glass transition at all compositions and optical transparency suggestive of miscible character. These optical adhesive blends showed zero birefringence or zero optical retardation at compositions between 60-70-wt% copolymer. The zero Birefringence afforded by the compensation of the intrinsic birefringence of the constituent polymer chains may be attributed to the complete miscibility of the blends in the cured state.

INTRODUCTION

Most polymers are intrinsically birefringent(optically anisotropic) due to the inherent asymmetry of polarizability of chemical bonds present in their long chain molecules. Depending on its chemical nature, a polymer can have a positive or negative inherent dielectric anisotropy. Birefringence can be induced. When a polymer is subjected to flow during extrusion and injection molding, the end product shows strong birefringence owing to frozen-in molecular orientation arising from residual and flow stresses.

Knowledge about birefringence is important in polymer research. Although residual birefringence is desirable for good mechanical properties as in fibers and films and for phase compensation as in polarizers and analyzers, residual birefringence could present undesirable effects to some optical applications such as optical disks and electronic display devices^{4,5}. Concerning about the problem of birefringence in polymers for optical and electro-optical uses, there has been serious efforts to reduce it to zero in recent years via three approaches: polymer blending, copolymerization, and fabrication process modifications⁶. This study employed the first two approaches in the development of non-birefringent optical adhesives.

The desired features of the proposed optical adhesives are complete miscibility at all compositions; glass transparency or optical clarity; curable or crosslinkable; adjustable

refractive index with composition, zero birefringence, excellent adhesive properties, and environmentally safe. These are highly stringent requirements.

Originally proposed system consisted of PMMA/EPON-828 blend system was chosen because of possible synergy of high optical clarity of PMMA and excellent adhesive properties of EPON-828. In a preliminary study, this system has been shown to phase separate when cured with TMA. As a result, a copolymer of MMA and GMA (MMA-co-GMA) was synthesized, and blended with EPON-828 then cured with TMA. The presence of GMA (with reactive epoxy groups) in the copolymer chain is to introduce functionality. Details of the phase separation study and of the copolymer synthesis and characterization are reported elsewhere², and are omitted here. This paper focuses on the formulation, characterization, and birefringence studies of TMA-cured MMA-co-GMA/EPON-828 optical adhesive blend systems.

Possible applications of the optical adhesives are in the bonding of optical devices without the distortion of the optical properties in the bonded areas as in optical disks and optical instruments and the bonding of visual display materials to other substrates as in electronic visual identification devices.

EXPERIMENTAL

Bis-phenol-A diglycidylether (EPON-828) prepolymer used in this study was purchased from Yuka Shell Company. It has 2 epoxy groups per molecule and epoxy equivalent of 380. The synthesized MMA-co-GMA copolymer used was made from a feed consisting of 90wt% MMA and 10wt% GMA. It has an average M_n of 122,099 with a polydispersity of 1.812.

The experimental work consisted of DSC for miscibility study and birefringence measurements. Five different blends were formulated in solution using acetone as common solvent. Each solution was of 10wt% concentration. Ten parts per hundred (10pph) of TMA was used for curing. For the DSC study, Dupont Thermal Analyzer Model 9900 was used at a heating rate of 10°C/min., while for the birefringent study, the instrument used was Metricon Prism Coupler Model 2010. Both smooth free standing films and samples cast on micro-glass slides were used with thickness range of 3-8µm. Curing was done between 150°C and 210°C.

RESULTS AND DISCUSSION

For all compositions, the formulated blends were transparent to the naked eyes both in solution and as cast or free standing films.

The second run DSC thermograms MMA-co-GMA blends without TMA curing is shown in figure 1. It is noticed quite clearly that a single T_g is obtained for each blend indicative of miscible character. These glass transitions are also observed to vary or shift in a defined manner with increasing copolymer composition.

For TMA-cured blends, figure 2 shows the second run DSC thermograms obtained. Again as in figure1, single T_g for each cured blend indicative of miscible character is

obtained. These glass transitions also vary systematically with compositions. However, higher glass transitions are obtained for the TMA-cured blends than for blends without TMA curing. This is due to the reduction in chains segmental motions as a results of crosslinking reaction which occurred. It is known the the effect of reduction in segmental of the chains is an increase in the glass transition

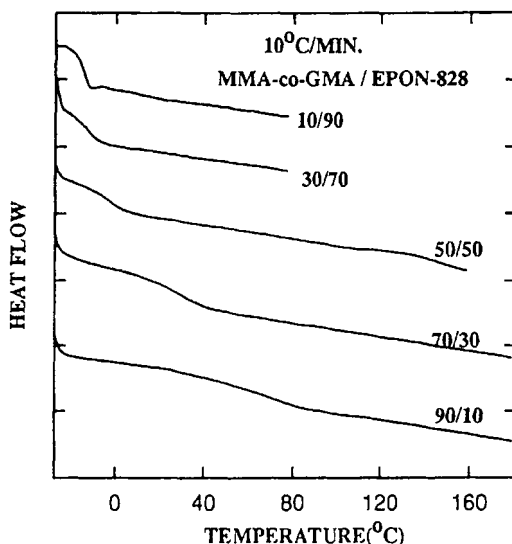


FIGURE 1 Second run differential scanning calorimetry thermograms of blends of MMA-co-GMA copolymer and EPON-828 without TMA curing.

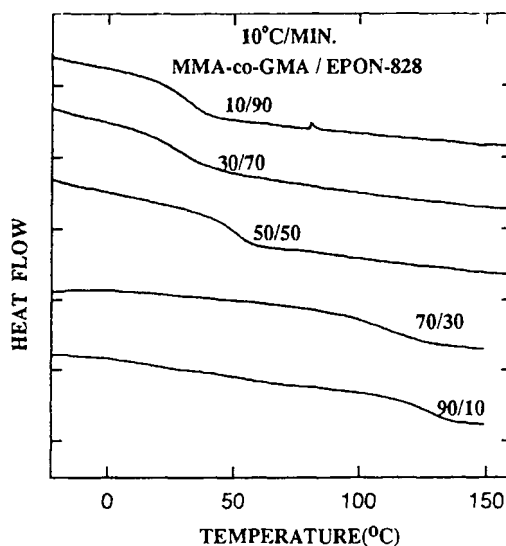


FIGURE 2 Second run differential scanning calorimetry thermogram of TMA-cured blends of MMA-co-GMA and EPON-828.

Finally, the birefringence of the TMA-cured blends represented as optical retardation or path difference is shown in figure 3 plotted against wt% copolymer. It is clearly observed that starting from a low amount of copolymer (high EPON-828), there is high optical retardation which decreases with increase in wt% copolymer, reaching a zero value between 60-70wt% copolymer. After this composition range, the negative value of retardation becomes larger with increasing wt% copolymer.

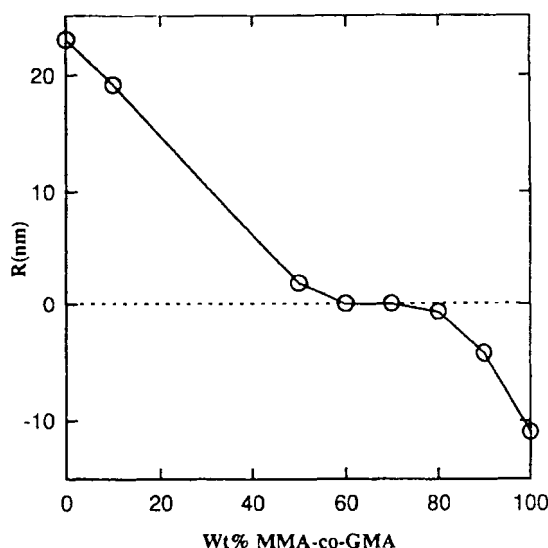


FIGURE 3 Plot of birefringence as optical retardation of TMA-cured blends of MMA-co-GMA copolymer and EPON-828 against wt% copolymer.

CONCLUSION

It has been possible to develop optical adhesive blends of MMA-co-GMA and EPON-828 cured with TMA. The optical adhesives were shown to be transparent and miscible at all compositions both in the uncured and TMA-cured states. It has also been possible to obtain zero birefringence for blends in the composition range 60-70wt% copolymer suitable for use in optical and electro-optical devices sensitive to residual birefringence.

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