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ABSTRACT (cont'd.)

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Final Report

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Contract N00014-77-C-0376

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June 1, 1977 - August 31, 1985

Phase Relations in Multicomponent Polymer Systems

R. J. Roe
Department of Materials Science
and Metallurgical Engineering
University of Cincinnati
Cincinnati, OH 45221-0012

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The research project under Contract N00014-77-C-0376 aimed at investigating polymer blends and in particular those blends containing random or block copolymers. The properties of polymer blends are determined by the degree of compatibility between the constituent polymers, and when they are incompatible, by the shapes and sizes of the domains they form and the degree of adhesion between them. In this project a number of polymer blend systems were studied to find the thermodynamic factors

controlling the compatibility, such as the temperature, concentration, molecular weight and the copolymer composition, etc. The specific topics investigated include the following:

- (1) The polymer-polymer interaction parameter between polystyrene and polybutadiene was determined, and the applicability of mean field theories (Flory-Huggins theory and the equation-of-state theory) to the polymer miscibility was ascertained.
- (2) The feasibility of determining the thickness of interfaces between dissimilar polymers by means of small-angle X-ray scattering was critically examined.
- (3) The occurrence of the order-disorder transition in block copolymer systems were investigated.
- (4) Light scattering and small-angle X-ray scattering were utilized to study the phase transition and phase separation behavior of mixtures of a homopolymer and a block copolymer, and this led to the construction of phase diagrams which exhibit many new features not found with other types of polymer blends.
- (5) Deuteration of polystyrene was found not to influence its miscibility with polybutadiene, in contradiction to what had been reported in the literature.
- (6) The effectiveness of a random or

block copolymer as a compatibilizer of homopolymer blends was investigated. ~~(7)~~ The effect of molecular weight polydispersity on the cloud point curves of polymeric blends was investigated. The results of these and other studies are described in the technical reports and publications as listed below.

The following personnel participated in the project:

- J. C. Chang, post-doctorate, 1977-1978
- M. Fishkis, post-doctorate, 1978-1981
- W. C. Zin, graduate student, 1977-1983
- D. Rigby, post-doctorate, 1981-1985
- L. Lu, graduate student, 1982-1984
- J. L. Lin, graduate student, 1983-1985.

List of Technical Reports Issued:

1. Evaluation of the Polymer-Polymer Interaction Parameter from the Equation-of-State Thermodynamic Theory, by R. J. Roe, June 1978.
2. Determination of the Polymer-Polymer Interaction Parameter for the Polystyrene-Polybutadiene Pair, by R. J. Roe and W. C. Zin, 1979.
3. Adaptation of a Kratky Camera to Use with a One-Dimensional Position-Sensitive Detector, by R. J. Roe, J. C. Chang, M. Fishkis, and J. J. Curro, August 1980.
4. Small-Angle X-ray Diffraction Study of Thermal Transition in Styrene-Butadiene Block Copolymers, by R. J. Roe, M. Fishkis, and J. C. Chang, 1980.
5. Examination of Errors in the Determination of Phase Boundary Thickness by Small-Angle X-ray Scattering, by R. J. Roe, June 1981.
6. Phase Equilibrium and Transition in Mixtures of a Homopolymer and a Block Copolymer. II. The Phase Diagram, by R. J. Roe and W. C. Zin, January 1983.
7. Phase Equilibrium and Transition in Mixtures of a Homopolymer and a Block Copolymer. I. Small-Angle X-ray Scattering Study, by W. C. Zin and R. J. Roe, March 1983.
8. Small-Angle X-ray Scattering Study of Micelle Formation in Mixtures of Butadiene Homopolymer and Styrene-Butadiene Block Copolymer, by D. Rigby and R. J. Roe, October 1983.
9. Effect of Polydispersity on the Cloud Point Curves of Polymer Mixtures, by R. J. Roe and L. Lu, June 1984.
10. Phase Diagram of Polymer Blends Containing Block Copolymers, by R. J. Roe, November 1984.
11. Deuterium Isotope Effect on the Compatibility between Polystyrene and Polybutadiene, by J. L. Lin, D. Rigby, and R. J. Roe, November 1984.

List of Publications Acknowledging ONR Support:

1. Roe, R. J., Evaluation of the Polymer-Polymer Interaction Parameter from the Equation-of-State Thermodynamic Theory, Adv. Chem. Series 176, 599 (1979).
2. Roe, R. J. and Zin, W.-C., Determination of the Polymer-Polymer Interaction Parameter for the Polystyrene-Polybutadiene Pair, Macromolecules 13, 1221 (1980).
3. Roe, R. J., Chang, J. C., Fishkis, M., and Curro, J. J., Adaptation of a Kratky Camera to Use with a One-Dimensional Position-Sensitive Detector, J. Appl. Cryst. 14, 129 (1981).
4. Roe, R. J., Fishkis, M., and Chang, J. C., Small-Angle X-ray Diffraction Study of Thermal Transition in Styrene-Butadiene Block Copolymers, Macromolecules 14, 1091 (1981).
5. Roe, R. J., Zin, W.-C., and Fishkis, M., Thermal Transitions in Polymer Mixtures Containing Block Copolymers, Proc. IUPAC Macro, Amherst, MA, p. 662, 1982.
6. Roe, R. J., Examination of Errors in the Determination of Phase Boundary Thickness by Small-Angle X-ray Scattering, J. Appl. Cryst. 15, 182 (1982).
7. Rigby, D. and Roe, R. J., SAXS Study of Micelle Formation in Mixtures of Butadiene Homopolymer and Styrene-Butadiene Block Copolymer, Polymer Preprints 24(2), 415 (1983).
8. Zin, W.-C. and Roe, R. J., Phase Equilibria and Transition in Mixtures of a Homopolymer and a Block Copolymer. 1. Small-Angle X-ray Scattering Study, Macromolecules 17, 183 (1984).
9. Roe, R. J. and Zin, W.-C., Phase Equilibria and Transition in Mixtures of a Homopolymer and a Block Copolymer. 2. Phase Diagram, Macromolecules 17, 189 (1984).
10. Rigby, D. and Roe, R. J., Small-Angle X-ray Scattering Study of Micelle Formation in Mixtures of Butadiene Homopolymer and Styrene-Butadiene Block Copolymer, Macromolecules 17, 1778 (1984).
11. Rigby, D. and Roe, R. J., SAXS Study of the Effects of Block Length on the Characteristics of Micelles Formed in Diblock Copolymer-Homopolymer Mixtures, Polymeric Materials Preprints 51(2), 382 (1984).
12. Roe, R. J. and Lu, L., Effect of Polydispersity on the Cloud-Point Curves of Polymer Mixtures, J. Polymer Sci., Polymer Phys. Ed. 23, 917 (1985).

13. Roe, R. J., Phase Diagram of Polymer Blends Containing Block Copolymers, *Polymer Eng. Sci.* 25, 0000 (1985).
14. Lin, J. L., Rigby, D., and Roe, R. J., Deuterium Isotope Effect on the Compatibility between Polystyrene and Polybutadiene, *Macromolecules* 18, 0000 (1985).
15. Rigby, D., Lin, J. L., and Roe, R. J., Compatibilizing Effect of Random or Block Copolymer Added to Binary Mixture of Homopolymers, *Macromolecules* 18, 0000 (1985).

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