Polymer Vol. 37 No. 12, pp. 2291–2297, 1996
Crown copyright © 1996 Published by Elsevier Science Ltd
Printed in Great Britain.
0032-3861/96/$15.00 + 0.00

polymer papers

Ambiguities in the interpretation of small-angle neutron scattering from blends of linear and branched polyethylene

C. Schipp, M. J. Hill* and P. J. Barham
H.H. Wills Physics Laboratory, Tyndall Avenue, Bristol BS8 1TL, UK

and V. M. Cloke, J. S. Higgins and L. Oiarzabal†
Department of Chemical Engineering, Imperial College, Prince Consort Road, London SW7 2BY, UK
(Received 8 December 1994; revised 5 April 1995)

This paper describes the difficulties of interpretation of small-angle neutron scattering data from blends of linear with lightly branched polyethylenes. The expected 'phase diagram' for the blend system was determined using indirect techniques, i.e. differential scanning calorimetry and transmission electron microscopy. Two different blends have been examined by neutron scattering at temperatures well above the melting point of both components. The neutron scattering data from one blend (mixed according to the 'phase diagram'), containing 50% linear deuterated material and 50% branched polymer, showed it to be demixed. The phase sizes stayed constant at around 23 nm over 36 h. The scattering from the other blend (demixed according to the 'phase diagram'), containing 10% linear deuterated material and 90% branched polymer, could be interpreted in two different ways. The data are consistent with a homogeneously mixed blend, but can equally well be interpreted as coming from a system which is phase-separated with a large domain size, as seen in the electron micrographs. Crown copyright © 1996 Published by Elsevier Science Ltd.

(Keywords: polyethylene blends; phase separation; neutron scattering)

INTRODUCTION

The question of whether blends of linear polyethylenes with lightly branched polyethylenes (polyethylenes with less than about 50 short branches per 1000 backbone carbon atoms) can exhibit any liquid–liquid phase separation in the melt has become somewhat controversial in the recent literature. Most of the experimental evidence for any phase separation comes from indirect experiments in which rapidly quenched blend samples are studied in the solid state. The problem with obtaining direct experimental evidence for, or against, phase separation lies in the close similarity in physical properties of the two components of such blends. In particular, the refractive indices of linear and lightly branched polyethylenes are so close to one another that no light scattering experiments are possible. At first sight, neutron scattering experiments, using blends of a linear deuterated polymer with a hydrogenous branched polymer, should provide a way to investigate phase separation directly in the melt. However, as we shall show, there can be considerable ambiguities in the interpretation of the data from such experiments.

Several authors have recently investigated polyethylene and blends of polyethylenes using techniques including neutron scattering[1–14]. Hydrogenous (HPE) and deuterated (DPE) polyethylenes are usually mixed together in various proportions and the scattering observed. The objectives have included determining the radii of gyration of the molecules, determining the χ parameter for interactions between HPE and DPE and studying the possibility of phase separation. In general any phase separation found has been interpreted as being due to isotope effects. Most of this work has only been concerned with linear polyethylenes.

In one particular paper[14] neutron scattering from blends of linear polyethylenes with lightly branched polyethylenes was studied. In this paper, Alamo *et al.* take issue with a body of work[15–33], mostly by ourselves, in which it has been argued, from indirect evidence, that such systems can undergo liquid–liquid phase separation in the melt at certain temperatures and blend compositions. Alamo *et al.*[14] suggested that their results demonstrated that the blends they studied were mixed in the melt at all compositions and temperatures, with any demixing being attributed to isotope effects introduced by the fact that the linear polyethylenes they used were deuterated.

In our earlier studies[15–20] we argued that the two distinct morphologies found in rapidly quenched blends of linear with lightly branched polyethylenes arose from some liquid–liquid phase separation in the melt prior to

* To whom correspondence should be addressed
† Present address: INASMET Centro Technologico de Materiales, Camino de Portuetxe 12, 20008 San Sebastian, Spain