

patterns similar to Figure 2 but the lithium salts are quite different, exhibiting no maximum and being very similar to the un-ionized acid copolymer. In addition, although annealing is known to increase the degree of crystallinity as measured by the wide angle X-ray technique,² it had very little or no effect on the low angle maximum in the cesium salt.

Conclusions

(1) The comparison between the scattering pattern obtained from the un-ionized acid copolymer and low-density polyethylene indicates that these two materials are structurally similar.

(2) The comparison between the scattering patterns obtained from the un-ionized acid copolymer and the cesium salt proves that there are fundamental structural differences between these two materials and hence that the cesium salt, in contrast to the un-ionized acid copolymer, possesses a structure different from that of low-density polyethylene.

(3) The maxima observed with cesium and sodium salts are unaffected by annealing which is known to alter the crystalline fraction. These maxima are, therefore, interpreted as being due to electron dense regions in the amorphous fraction which are almost certainly caused by aggregations of ions. The fact that no maximum is observed with the lithium salt supports this view inasmuch as lithium scatters X-rays very weakly. However, the similarity between the mechanical and viscous properties of the lithium salt and those of other salts indicates structural similarities between them.

(4) Therefore, the preliminary low angle X-ray results provide direct evidence for the three-phase structural model for the ionized copolymers. Further work is in progress and will be described in due course.

Experimental Section

The starting copolymer was kindly provided by the Du Pont Co. It was stated to have a weight average molecular weight of 300,000 and oxygen analysis established the presence of 4.1 mol % methacrylic acid units. The finely divided starting material was refluxed in tetrahydrofuran with hydroxides of lithium, sodium, and cesium in order to produce the ionized copolymers. Infrared analysis established that the following ionizations were obtained: Li salt, 65%; Na salt, 60%; Cs salt, 59%.

After ionization the samples were precipitated twice in a methanol-water mixture, washed thoroughly several times, and dried *in vacuo* at 100°. Samples were compression molded for X-ray analysis. Annealing was carried out by heating the samples at 10° below their melting points for 24 hr under vacuum, followed by slow cooling to room temperature.

Low angle X-ray scattering patterns were obtained using a Warhus low-angle camera with 0.020-in. pin holes and a 29-cm sample-to-film distance. The camera was evacuated during exposures and the scattering patterns, which were all obtained with nickel-filtered copper radiation, were microdensito-metered using a Joyce-Lobel instrument.

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Page 137. In the last sentence of the subsection entitled Description of Model and Notation N_0 should be redefined as a molar quantity.

Page 138. Equation 2 is incorrect. It should read

$$\Delta S_D/N_0 = -R \{ X_A [(1 - P_{AA}) \ln (1 - P_{AA}) + P_{AA} \ln P_{AA}] + X_B [(1 - P_{BB}) \ln (1 - P_{BB}) + P_{BB} \ln P_{BB}] \}$$

Page 141. The parentheses in the subscript in the upper right hand corner of Figure 4 should be eliminated.

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