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Morphological Origin of Thermomechanical Behavior in Semicrystalline Ethylene/Methacrylic Acid Ionomers
KATSUYUKI WAKABAYASHI, RICHARD A. REGISTER, Department of Chemical Engineering, Princeton University — Two peculiar and intriguing phenomena in ethylene/methacrylic acid (E/MAA) ionomers are an initial sharp increase in stiffness with neutralization and an inverse dependence of Young's modulus on crystallinity. We have identified how the polyethylene crystallites, amorphous polymer segments, and ionic aggregates combine to produce these unusual effects. At temperatures just below the melting point of the primary crystallites, the ionomers can be satisfactorily described as two-phase composites of crystallites and ionically-crosslinked rubber, but at room temperature, the modulus is much greater. We trace this effect to a synergy between the ionic aggregates and secondary crystallites, which together form percolated rigid pathways through the amorphous phase at room temperature, generating a far higher modulus than one would anticipate from the modest crystallinity and ion content. When the secondary crystallites melt and/or the segments in the regions of restricted mobility surrounding the aggregates devitrify, these paths break down and the simple two-phase composite description is recovered.

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