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Influence of Neutralization on Amorphous-Phase Properties in Semicrystalline Ionomers KATSUYUKI WAKABAYASHI, RICHARD A. REGISTER, Department of Chemical Engineering, Princeton University, Princeton, NJ 08544-5263 — Ethylene-methacrylic acid (E-MAA) ionomers contain lamellar polyethylene crystallites, amorphous copolymer segments and ionic aggregates, each of which affects the mechanical properties of the material. For a quantitative assessment of the contributions from each of the three structural motifs, we measured the ionomer modulus at 70 °C, where the materials still contain substantial crystallinity, and applied a two-phase composite treatment (Davies Model) to extract the modulus of the amorphous phase. The amorphous phase modulus at 70 °C increases with neutralization level as a consequence of physical crosslinking by the ionic aggregates; amorphous phase moduli for ionomers with varying comonomer content and neutralization levels approximately collapsed when plotted against the number density of ionic groups, with the modulus increasing with ion content in general agreement with simple rubber elasticity theory. Between 25 and 70 °C, the relaxation behavior of ionomers differs substantially from that for unneutralized E-MAA copolymers. The ionomers exhibit two-step drops in the storage modulus prior to primary crystal melting, which we attribute to melting of secondary crystallites and devitrification of the amorphous phase, whose glass transition is elevated by neutralization.

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