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Linear Viscoelastic and dielectric behavior of Phosphonium Ionomers QUAN CHEN, SIWEI LIANG, U HYEOK CHOI, JAMES RUNT, RALPH H. COLBY, Department of Materials Science and Engineering, The Pennsylvania State University — Linear viscoelastic (LVE) and dielectric (DRS) responses were examined for polysiloxane-based phosphonium-ions with fractions of ionic monomers $f = 0$ to 0.3 ; the other monomers have short poly(ethylene oxide) side chains. LVE of these samples shows a glassy relaxation followed by a terminal polymer relaxation that is increasingly delayed with increase of f . The glassy relaxation broadens when $f > 0.1$. DRS of these samples shows a segmental α process associated with motion of monomers, followed by an additional $\sim 100X$ slower α_2 process before electrode polarization. A detailed comparison between LVE and DRS reveals that the α_2 relaxation in DRS corresponds to a characteristic modulus of $k_B T$ per ionic group in LVE. This result strongly suggests that the molecular origin of the α_2 relaxation is the dissociation/association of the ionic groups from/into the ionic clusters, consistent with the observed magnitude of the α_2 relaxation increasing with ion content. Based on this molecular view, we can predict the terminal polymer relaxation from the α_2 relaxation time obtained in DRS, assuming this is the lifetime of ionic associations in a sticky Rouse model. Meanwhile, the broadening of glassy mode distribution with increasing $f > 0.1$ is attributed to an enhanced cooperation for motion of glassy segments. This enhancement is possibly due to decrease of distance between the ionic groups with increasing f , leading to stronger overlap of polarizability volumes.

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