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Effect of Chemical Structure on the Depth and Temperature Dependence of Dynamics Near the Free Surface of Polymer Glasses

The dynamics of polymer glasses have been shown to be enhanced within a few nanometers of an interface. This enhanced surface mobility has technological implications for processing and applications of glasses. An open question is how the chemical structure of a polymer affects the dynamics in the near surface region, but this is extremely challenging to study. Previous studies using bulky fluorescent probes found a mobile surface layer in polystyrene (PS) but not in poly(α-methylstyrene) (PαMS). We have used βNMR to study the depth dependence of phenyl ring dynamics in PS (T_g = 370~K) and PαMS (T_g = 448~K) glasses at 317 and 261~K. The larger T_g of PαMS indicates the polymer chains are stiffer than PS. The βNMR results indicate there is a mobile surface layer in PαMS contrary to previous reports and that this layer is much thicker than that of PS. The characteristic length scale for the return to bulk dynamics at 317~K is 26(3)~nm in PαMS versus 4.0(2)~nm in PS. The stiffness of the polymer chains plays a role in the transmission of fast surface dynamics into the bulk. The phenyl ring motion appears to be decoupled from the backbone motion.

Email

iainmckenzie@triumf.ca

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Supervisors Name

Supervisors Email

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Primary author: MCKENZIE, Iain (TRIUMF)

Co-authors: MACFARLANE, Andrew (UBC Chemistry); Dr MORRIS, Gerald (TRIUMF); Prof. FORREST, James (University of Waterloo); TICKNOR, John O. (Department of Chemistry, University of British Columbia); Ms MEHMOOD, Mahnoor (University of Waterloo); LI, Ruohong (TRIUMF); KARNER, Victoria L. (TRIUMF)

Presenter: MCKENZIE, Iain (TRIUMF)

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