Chapter 6

Conclusion and Future perspectives

A fundamental question in glassy physics is the relationship between the structure and properties of glassy systems and to understand the origin of glass transition phenomenon. The study of glass transition phenomenon using Raman spectroscopy in this thesis provides evidence of the molecular origin of the glass transition phenomenon. Raman spectroscopy being a vibrational spectroscopy, provides insight into the molecular orientations and helps in tracking configurational changes and molecular mobility during the thermal cycling of a polymer films. This establishes a connection between the glassy dynamics and the molecular motions in the polymer. The correlation between molecular mobility obtained from Raman peaks and the tensile properties also establishes the interrelation between the molecular vibrations and macroscopic mechanical properties. This highlights the significance of the study molecular mechanisms controlling the macroscopic properties.

The study of response to thermal perturbation in glassy polymer shows that the relaxation behavior of a polymer films show a transition from the caging dynamics to cooperative segmental dynamics on crossing the glass transition temperature. The work presented here also provides evidence to the presence of cooperatively rearranging regions, as explained by the random first order theory of glasses. The presence of dynamic heterogeneities in a 2-dimensional mapping of Raman modes in glassy state of polymer film establishes an important conclusion that the glass transition phenomenon is accompanied with the dynamic heterogeneities not only at the microscopic level but also at the molecular level.

Another important outcome of the present study is the study of micro-scale mechanical properties which emphasizes the importance of molecular interactions which have an impact on the structure of the polymer matrix, and

micro-scale diffusion dynamics. The study also provides a methodology to use the FRAP technique to study micro-mechanical properties and a simultaneous structural analysis on addition of filler particles in the polymer matrix from FRAP pre-bleach image data. The current work has studied the effect of micro structuring like aggregation due to the presence of interactions within the polymer matrix on the macroscopic mechanical properties. It is established by the present studies that for better applicability of any polymer and for the development of new polymer-based products, it is important to study the molecular as well as microscopic properties in order to achieve desirable macroscopic properties. Overall, this study lays out the importance of looking at different length scales to get an in depth understanding of glassy dynamics.

The study attempts to provide an experimental evidence to some fundamental questions in glassy physics but there are few questions that need to be addressed like, is the glassy dynamics is universal for all the polymer in all the geometries? If the glassy dynamics has a structural correlation? The need of the hour is development of a model which can provide a better understanding of the glass transition phenomenon and its universality. Also, development of polymer nanocomposites with tunable properties is becoming popular due to advancing applications of polymers but a thorough understanding of molecular as well as microscopic mechanisms of nancomposites before their use in various applications. The glassy dynamics is often known to alter upon confinement therefore, it is also important to study different confinement geometries of different polymers for a better understanding of glassy dynamics in polymer films. Last but not the least, development of a biodegradable polymer which can be replace the widely-used non-biodegradable polymers is necessary.