Thursday,

10th January 2013

at: **5.15 pm**

Gustav-Mie-Hörsaal, Theodor-Lieser-Str. 9, 06120 Halle

Coffee will be served from 4.45 pm!

Prof. Dr. Juan COLMENERO

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Constrained Chain dynamics in un-entangled polymer blends with dynamic asymmetry. A generalized Langevin equation (GLE) formalism

Recent molecular dynamics (MD) simulations in both bead-spring models [1] and atomistic blends [2] as well as different experimental investigations have put forward evidence that the chain dynamics of linear un-entangled polymers are drastically modified when they form part of dynamically asymmetric polymer blends. These are miscible polymer systems where the two components display very different mobility (or glass transitions temperatures). MD simulations show that this dynamic asymmetry between the two components induces strong non-exponentiality of the Rouse mode correlators corresponding to the fast component of the blend. This non-exponentiality is a signature of the breaking of one of the main assumptions of the Rouse model: *the time un-correlation of the external forces acting on the tagged chain*. Moreover, a scaling $t_p \sim (N/p)^X$ was reported [1], where t_p is the relaxation time corresponding to the p-Rouse correlator and N the number of "beads" of the chain. The parameter X, strongly depends on the temperature, T, varying from $X \approx 2$ (Rouse prediction) at high T – where the dynamic contrast between the two blend components is very small – to $X \approx 3.5$ at rather low T, where due to the slowing down of the slow component, the fast component moves in an almost "frozen matrix". These simulation results, obtained for a generic bead-spring model, suggest that the molecular weight dependence of the Rouse time t_R (t_p; p=1) should also depend on temperature and be given by $t_R \sim N^X$. This behavior has been recently confirmed by dielectric spectroscopy in asymmetric blends of polyisoprene with different molecular weights and poly(tert-butylstyrene) [3].

The abovementioned results can be understood in a theoretical framework based on the generalized Langevin equation (GLE) formalism. In this framework, a direct and nontrivial correlation between the X(T) parameter giving the wave length (N/p) dependence of the relaxation times t_p and the non-exponential parameter of the corresponding Rouse mode correlator has been deduced [3]. This correlation is nicely corroborated by the MD simulation results. Moreover, a generalized Rouse incoherent scattering function can also be deduced with some approximations within the GLE formalism [4]. The obtained expression reduces to the well know Rouse incoherent scattering function (de Gennes) in the limit of pure Rouse behavior. The validity of the expression found is checked by fully atomistic MD simulations on poly(ethylene oxide)-poly(methyl methacrylate) blends [2], which were validated by neutron scattering [5].

[1] A. Moreno and J. Colmenero, Phys. Rev. Lett., 100, 126001 (2008).;[2] M. Brodeck, F. Alvarez, A. J. Moreno, J. Colmenero, D. Richter, Macromolecules, 43; [3] S. Arrese-Igor, A. Alegría, A. J. Moreno and J. Colmenero, Soft Matter, 8, 3739 (2012);[4] J. Colmenero (to be submitted); [5] M. Brodeck, F. Alvarez, J. Colmenero, D. Richter, Macromolecules, 45, 536 (2012).3036 (2010).

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