Understanding the viscoelastic behavior of linear telechelic polymers governed by both transient bond dynamics and interchain entanglements

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We study the rheological behavior of linear metallo-supramolecular polymers based on hydrogenated terpyridine-terminated polybutadiene polymer chains with different metal salts Since our system is composed of a non-polar polymer and a polar end groups, partial phase separation of the end-groups is expected, in addition to the formation of long linear assemblies through metal-complex coordination. The linear viscoelastic behavior of these chains shows two plateaus. The relaxation following the first plateau is attributed to the relaxation of the linear assemblies free to move, while the second, low frequency plateau is attributed to the molecular segments trapped into the network generated by the phase separation of some metal-ligand complexes. These trapped segments can only partially relax by Constraint Release Rouse motion exploring their surrounding at the rhythm of the entanglement/disentanglement of the free chains and dangling ends. Then, a slow relaxation process appears at low frequency, which could be due to several mechanisms. We attribute it to the breaking of the complexes along the self-assemblies. We then test this molecular picture with the help of a tube model.