



Dynamical Relaxations of Cross Linked Networks in Polymer Solutions

Mustapha Benmouna and Farida Benmouna*

Macromolecular Research Laboratory, Faculty of Sciences, University of Tlemcen, Algeria

*Corresponding author: Farida Benmouna, Macromolecular Research Laboratory, Faculty of Sciences, University of Tlemcen, BP 119, Algeria, Tel +213771917486; E-mail: fbenmouna@email.com

Received: December 05, 2016; Accepted: December 09, 2016; Published: December 12, 2016

Introduction

Cross linked polymer networks and gels have been the subject of a particular attention in the literature for decades not only because of their fundamental interest but also because of their applications [1]. The presence of crosslinks leads to a physical behavior differing from that of pure free chain systems due to interplay between hard and soft properties. They combine the elasticity of polymer chains and the solidity of the assembly of strands holding together with covalent bonds. They can absorb large amounts of a low molecular weight good solvent (water in the case of hydrogels) undergoing substantial swelling. They also contract by expelling the solvent under certain conditions. Contraction and swelling are governed primarily by the nature of constituents in the medium, environmental conditions or external perturbations. The crosslinking density determines the mean size of the polymer strand between consecutive crosslinks and plays a major role in the solvent uptake or degree of de-swelling. In addition to low molecular weight solvents, networks have the ability to sequester macromolecular species such as DNA, proteins and enzymes. This property has extended the area of interest of cross linked networks and opened up new opportunities triggering intensive research work in recent years [1]. Most efforts focus on attempts to elucidate the impact of relevant parameters and multicomponent interactions within the system. Gene therapy and drug delivery based on encapsulated DNA, protein or drugs, highly sensitive smart devices are among the applications where one needs to understand the behavior of cross linked networks embedded in macromolecular solutions. Diffusion of macromolecules in and out of the network depends crucially on the degree of swelling and the size of macromolecules compared to the strand. The rate of macromolecular release may be tuned with the swelling kinetics. Hence, one of the main challenges in the controlled drug release is to identify the parameters and conditions that govern the release of macromolecules from the network at a given time rate in a specific targeted location. This is of primary importance to improve the efficiency of treatment and reduce undesirable collateral effects.

Dynamics of Cross Linked Networks Embedded in Polymer Solutions

The kinetics of swelling and de swelling (or contraction) of the networks in a low molecular weight solvent is governed by a fast process of solvent transport in or out of the network preceding a much slower relaxation process of polymer strands between consecutive crosslinks. If linear chains are present in the solution, the dynamics are more complicated because of interdependence of network relaxations and linear chain diffusion [1]. In an upcoming work we intend to report results in an effort to elucidate those coupled dynamical processes and shed light on the parameters controlling the collective diffusion of the system as well as the inter diffusive of one species with respect to the other according to their individual characteristics and mutual interactions. The dynamical relaxations of crosslinked networks embedded in polymer solutions will be modelled using linear response theory combined with generalized random phase approximation formalism. Recently, we reported an investigation of similar systems focussing on the phase behavior and structural properties [2-3]. We intend to extend this investigation to dynamical properties and relaxation modes. The intermediate scattering function is analyzed in terms of fast

and slow modes under a variety of conditions. Effects of the polymer composition, interaction parameters, size of the free chains and network strands, mismatch of solvent quality with respect to both polymer species is scrutinized in details.

Physical Interpretation of the Relaxation Eigen-Modes

The fast relaxation is attributed to the network breathing mechanism while the slow relaxation is reminiscent of the interdiffusive process and diffusion of linear chains in the network. This means that the fast mode is overwhelmingly governed by the network parameters while the slow mode relies more on the linear chain dynamics. The results are analyzed in the Rouse-Zimm limits with a special focus on the effects of long range hydrodynamic interactions. This study may be relevant to drug delivery and drug controlled release systems as well as protein partitioning in polymer networks.

References

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