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Molecular Dynamics Study on the Aggregation Behavior of Triton X Micelles with Different PEO Chain Lengths in Aqueous Solution

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Abstract: The aggregation structure of Triton X (TX) amphiphilic molecules in aqueous solution plays an important role in determining the various properties and applications of surfactant solutions. In this paper, the properties of micelles formed by TX-5, TX-114, and TX-100 molecules with different poly(ethylene oxide) (PEO) chain lengths in TX series of nonionic surfactants were studied via molecular dynamics (MD) simulation. The structural characteristics of three micelles were analyzed at the molecular level, including the shape and size of micelles, the solvent accessible surface area, the radial distribution function, the micelle configuration, and the hydration numbers. With the increase of PEO chain length, the micelle size and solvent accessible surface area also increase. The distribution probability of the polar head oxygen atoms on the surface of the TX-100 micelle is higher than that in the TX-5 or TX-114 micelle. In particular, the tail quaternary carbon atoms in the hydrophobic region are mainly located at the micelle exterior. For TX-5, TX-114, and TX-100 micelles, the interactions between micelles and water molecules are also quite different. These structures and comparisons at the molecular level contribute to the further understanding of the aggregation and applications of TX series surfactants.

Keywords: Triton X; micelle; hydration number; solvent accessible surface area

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1. Introduction

Micelles aggregated from surfactant molecules have been widely used owing to their ability to dissolve hydrophobic compounds effectively. People's interest in micelle solutions comes from their application potential as functionally molecular assemblies [1]. Different cationic, anionic, zwitterionic, and non-ionic surfactants have been widely studied. Compared with ionic surfactants, non-ionic surfactants are much less toxic and have more efficient surface active properties [2]. Among them, the series of Triton X (TX) non-ionic surfactants have been studied in depth and characterized [3]. Because of their unique molecular structure and amphiphilic nature, the micelles formed by TX surfactants have very flexible surface-active properties. The critical micelle concentration (CMC) of TX surfactant is very low, especially in aqueous solution. Different TX surfactants have been widely used in the fields of microbiology and biomedicine [4].

The TX-100 as one typical nonionic surfactant [5–7], has been extensively studied [8–10]. This surfactant consists of one hydrophilic chain of 9–10 ethylene oxide units linked to a benzene ring with an octyl chain [6]. Many researchers have explored the structural characteristics of TX-100 micelles in aqueous solution and their interaction with water by experimental techniques such as 2D NOESY NMR [11–13], pulsed field gradient NMR [8], solvent paramagnetic relaxation enhancement [7], fluorescence spectra [14,15], surface tension [15], light scattering [16], static and dynamic light scattering [15,17], turbidimetric method [18], small-angle X-ray scattering [13], quasi-elastic light scattering spectroscopy [19]. It is very

Molecular Dynamics And Aggregation Behavior In Aqueous Polymerdrug Model Systems

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Molecular Dynamics and Aggregation Behavior in Aqueous Polymer-Drug Model Systems Hans Evertsson,1999 *Acta Universitatis Upsaliensis* ,1999 *Polymer Preprints, Japan* ,2000 Index to Theses with Abstracts Accepted for Higher Degrees by the Universities of Great Britain and Ireland and the Council for National Academic Awards ,1993 Theses on any subject submitted by the academic libraries in the UK and Ireland SPSJ ... Annual Meeting Kōbunshi Gakkai (Japan),Kōbunshi Gakkai (Japan).,2000 **Process and Chemical Engineering** ,1998 Structure and Molecular Dynamics in Polymer Model Systems Abdallah Merenga Sarroney,1999 *Backbone Connectivity and Collective Aggregation Phenomena in Polymer Systems* Wen-Jong Ma,Chin-Kun Hu,2012 Modeling by the Method of Molecular Dynamics of Polymer Systems Oriented with the Aid of an External Field M. N. Lukyanov,A. A. Darinskii,Y. Y. Gotlib,I. M. Neelov,1985 *Molecular Dynamics Study of a Water Soluble, Upper Critical Solution Temperature Polymer* Yang Zhou,2014 Thermo responsive polymers have been the subject of research in academic and applied science over the past few decades Among them water soluble thermo responsive polymers draw most attention because water is the most common solvent for inorganic and living systems The key property of thermo responsive polymers is the change of its solubility in water due to change of temperature Using molecular dynamics simulations with different force fields water models and partial atomic charges assignment methods the upper critical solution temperature polymer in water was investigated To investigate the molecular mechanism for the UCST behavior molecular dynamic simulation was used along with poly n acryloyl glycinamide poly NAGA a non ionic homopolymer exhibiting the UCST behavior **Corpvs vasorvm antiquorvm** ,1968 **Molecular Dynamics Investigation on the Aggregation of Polyaromatic Compounds in Water and Organic Solvents** Cuiying Jian,2015 Aggregation of polyaromatic PA compounds has drawn great interest due to their wide impacts in areas such as petroleum processing Despite the extensive studies on PA compounds fundamental knowledge of their aggregation behaviors is still missing at atomistic level For instance it s still unclear how the properties of the solvent can lead to different aggregation mechanisms and hence affect the aggregated structures of PA molecules In this dissertation a series of molecular dynamics MD simulations have been performed to investigate the effect of solute chemical structures as well as solvents on the aggregation of PA compounds The PA molecules studied here possess the same PA core structure but have systematically varied side chain lengths We started with simulating a single type of PA compounds in water Inside the aggregated structures while some PA core stacking was observed most of the PA molecules are simply entangled together without preferred orientations More interestingly it was found that side chain length has a non monotonic effect on the size of the aggregates with intermediate side chain length leading to smaller aggregates In contrast regardless of the side chain length these PA molecules aggregated into ordered structures in toluene and n heptane which mainly consist of stacked PA cores On the other hand the ranges of stacking of PA cores in these two organic solvents are different thus resulting in

distinct aggregated geometries Following the above studies which involve only a single type of PA compounds as the solute we explored the aggregation of mixed PA compounds of different types in toluene n heptane and heptol toluene n heptane mixture It was found that the inhomogeneity in solutes can enhance the stacking of PA cores leading to the long range stacking of PA cores Furthermore the existence of this long range stacking of PA cores is insensitive to the solvents employed Through detailed analysis of the aggregated structures the aggregation mechanisms of different kinds of solutes were clarified This dissertation provided insights for the aggregation of PA compounds from atomic level and shed lights on controlling their aggregated structures **Molecular Dynamics Investigations of Polymer Solutions Undergoing Shear Flow** Carsten Aust,2000 Accelerated Molecular Dynamics Simulation Method Development and Applications in Polymer Systems Modan Liu,2021* **Structure and Dynamics of Associative Polymer Gels** Ameya Rao,2023 Associative polymers have attracted wide attention for applications spanning biomedicine soft robotics and rheological modification due to their unique viscoelastic and stimuli responsive properties imparted by their dynamic bonds However predicting the response of associative polymer networks remains a challenge due to the interplay between chain and bond dynamics on various timescales which governs their properties such as self diffusion relaxation and creep In this thesis a combined experimental and computation approach was used to provide new insight into the structure and dynamics of associative networks on various length and time scales with the goal of understanding effects of key molecular parameters on their macroscopic behavior The first part of this thesis studied molecular relaxation and self diffusion of a model associative network formed by artificial coiled coil proteins with a well defined architecture The combination of forced Rayleigh scattering FRS and neutron spin echo NSE spectroscopy provided evidence for several regimes of gel relaxation behavior across a wide length scale range including subdiffusive caging and two distinct regimes of apparent superdiffusion before terminal Fickian diffusion The submolecular relaxation dynamics were further probed by varying the strand length and chain concentration illustrating changes in segmental motion that reflected the underlying network design Importantly segmental relaxation rates were found to collapse onto a master curve when rescaled by the static inter junction spacing measured by neutron scattering indicating self similar dynamics even in networks with different chain architecture and concentration Furthermore the presence of two distinct superdiffusive regimes on intermediate length scales suggested the existence of multiple origins for anomalous diffusion in associative systems reflecting a complex interplay between distinct molecular states not captured by current theories To obtain further insight into the molecular mechanisms underlying associative network dynamics a coarse grained bead spring model was developed and implemented via Brownian dynamics simulations Associative polymers were conceptualized as linear chains containing regularly spaced stickers interacting with a mean field background obviating the need to explicitly model multi chain interactions and reducing computational cost The simulations demonstrated the coexistence of multiple diffusive modes termed walking and hopping that give rise to the superdiffusive

behavior seen experimentally Importantly the two superdiffusive regimes were found to occur by distinct mechanisms with the lower regime occurring due to a transition between multiple walking modes irrespective of the hopping mode and the upper regime occurring due the onset of molecular hopping Molecular hopping was shown to be important only in kinetics limited systems where the sticker association dissociation dynamics are slower than the intrinsic Rouse relaxation of the network strands The simulations were also used to probe the effects of molecular parameters such as the sticker density chain concentration and association dissociation kinetics on self diffusion Notably the results demonstrated the importance of loops in enabling chains with high sticker density to hop whereas the mean field prediction of purely Fickian diffusion of all length scales was recovered at high chain concentration where the hopping mode was effectively suppressed Analytical theories were formulated to predict the characteristic walking and hopping diffusivities from the chain topological statistics and dissociation association timescales finding qualitative agreement with simulation Finally the last part of the thesis explored the possible contribution of multi chain correlations toward network dynamics on different length scales effects that are not captured by single chain conceptualizations commonly used Structural characterization of a model associative protein gel using small angle and ultra small angle neutron scattering provided evidence for a previously unobserved static correlation length larger than the inter junction spacing indicating inhomogeneity in the chain density distribution in the gel Self diffusion measurements suggested a caging effect induced by this large scale correlation length in governing a transition between distinct slow and fast diffusive modes Finally a comparison to the single sticker dissociation time inferred from tracer diffusion measurements supported the single chain mechanisms of walking and hopping as previously conceptualized with the step size of the slow mode commensurate with the length of the bridging strands and the transition timescale to the fast mode consistent with the onset of hopping via dissociation of all stickers on a chain A Molecular Dynamics Study of the Influence of Chain Branching on the Properties of Polymer Systems Martin Oliver Steinhauser,2001 **Effects of Nanoscale Aggregation on Mechanical Properties and Local Dynamics of Precise Acid- and Ion-containing Polymers** Luri Robert Middleton,2016 The structural regularity of precise polyethylenes also enables robust comparisons between experiments and computer simulations At pico to nano seconds time scales and length scales of polymer and aggregate dynamics neutron scattering and molecular dynamics simulations were combined to extend the knowledge of the molecular level aggregated polymer dynamics These experiments provide a baseline for future studies of ion conduction in associating polymer melts Molecular Dynamics Simulation of Polyatomic Molecules in Aqueous Solution Olle Teleman,1986 Molecular Dynamics Simulations of Polymer Liquids on Substrates of Different Topography Nikita Tretyakov,2012 The properties of polymer films and droplets at substrates of different topography are studied employing particle based simulation techniques Molecular Dynamics The liquid is modeled by short coarse grained polymer chains of 10 monomers while the temperature of the system is controlled by the Dissipative Particle Dynamics DPD thermostat that

conserves momentum locally and provides correct hydrodynamics Throughout this dissertation we show that macroscopic concepts cannot be straightforwardly extrapolated down to microscopic systems At first the flat topography of the substrate is st

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