MODELING THE SHEAR AND EXTENSIONAL RHEOLOGY OF SALIVA AND MUCIN HYDROGELS USING A STICKY GEL NETWORK MODEL

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There is increasing interest in using rheological measurements of saliva and other bodily secretions such as cervical and respiratory mucus as non-invasive diagnostics for pathology and disease. However, there is only limited literature available on the shear and extensional rheology of saliva, and nearly no consideration of its temporal stability in the face of biological degradation. Indeed, capillary breakup extensional rheometry (CaBER) data of saliva samples at various ages shows that both the time to breakup and relaxation time of these highly elastic but low viscosity aqueous solutions decrease as a function of age. The viscoelastic properties of saliva can primarily be attributed to the presence of large glycoproteins (MUC5B mucins) in solution. It is well known that these 'MUCmers' physically associate and interact with each other and their surroundings via ion-mediated crosslinking and hydrogen bonding interactions to form a weak hydrogel or 'pre-gel'. This motivates the development of a Sticky Network model for mucin-containing solutions, building on earlier work of Tripathi et al for synthetic HEUR associative polymer systems. The mucin macromolecules are modeled as a semi-dilute and semi-flexible network of physically-associated finitely extensible elastic segments with a stretch-dependent 'stickiness' energy parameter that must be overcome in order for the chains to be able to reversibly dissociate from the rest of the network under imposed deformation. We show that this model is able to accurately capture capillary thinning and filament rupture behaviour of saliva using biologically-derived parameters, and can systematically account for temporal changes in the rheology through a progressive decrease in the molecular weight of the MUC5B chains. To probe the role of the different association mechanisms in the network we construct a series of model mucopolysaccharide hydrogels using purified MUC2 and MUC5 mucins in aqueous solution, We characterize the linear and nonlinear rheology of these physically-associated networks and show that their linear viscoelastic properties are characterized by broad power-law relaxation characteristics. To demonstrate the very wide range of time-scales and length-scales that characterize the different network interactions present in these hydrogels we use a series of additives (low-molecular weight surfactants, salt and reducing agents such as acetylcysteine) to respectively disrupt the hydrophobic, ionic and disulphide interactions between the MUCmers that form the hydrogel network.