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Nonlinear Elasticity of Bottlebrush Networks and Gels¹ ANDREY DOBRYNIN, ZHEN CAO, University of Connecticut, JAN-MICHAEL CAR-RILLO, Oak Ridge National Laboratory, SERGEI SHEIKO, University of North Carolina at Chapel Hill — Bottlebrush networks are examples of supersoft elastic materials that demonstrate highly nonlinear stress-strain behavior leading to material hardening with increasing deformation. Using molecular dynamics simulations and theoretical analysis we studied correlations between mechanical properties of bottlebrush networks and molecular parameters. Our simulations showed that both the network shear modulus G and the elongation at break decrease (onset of finite extensibility) with increasing the degree of polymerization (DP) of the side chains. The finite extensibility behavior is ascribed to the increase of the backbone elongation ratio β with DP of the side chains. Simulation results are in a good agreement with experimental observation of progressive softening of bottlebrush elastomers with increasing length of side chains and predictions of the nonlinear network deformation model which provides universal relationship between nonlinear network deformation modulus as a function of the first deformation invariant I1, bottlebrush backbone elongation ratio β , bottlebrush effective bending constant K and concentration of crosslinks.

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