SUPPORTING MATERIAL

Rate-dependent behavior of the amorphous phase of spider dragline silk

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Effect of the system size on friction forces

Fig. S1 shows the total friction forces per residue between the peptide chains of the amorphous phase at different pulling velocities. We have considered three simulation systems of bundles composed of 4, 8 and 24 chains to assess the system size dependency of friction forces. For the 4-chain bundle, the total friction forces per residue were low compared to the other two simulation systems, because in this simulation system only two chains were pulled in each direction, so that they were only minimally surrounded by chains pulled in opposite direction. For the 8-chain and 24-chain bundles, the total friction forces per residue were within the range of standard error. Therefore, friction forces can be considered to be independent beyond an 8-chain bundle, and thus, all our simulations were carried out for the 8-chain bundle simulation system, which was computationally more feasible than the 24-chain bundle, in particular at low pulling velocities.

Calculation of the velocity gradient parameter

We calculated the coefficient of viscosity η for the amorphous phase of dragline silk from our MD simulations data by using Newton's law of shear viscosity. $\tau = \eta \times dv/dx$, where τ is the shear stress, and dv/dx is the shear velocity or velocity gradient. To calculate dx, the velocity gradient parameter, we have considered our simulation data for the fastest velocity, which was 100 m/s. The computed dynamic viscosity of water $\sim 0.8\times10^{-3}$ Ns/m^2 was considered for the calculation. The shear stress (τ) is defined as, $\tau = F/A$, where F is the friction force, and A is the contact area. From the analysis of the simulation data, with F = 6921 pN, A = 6.3 nm^2, and η = 0.8×10^{-3} Ns/m^2, we obtained an effective length of dx = 10 nm.

Calculation of shear modulus

We considered two options to calculate the shear modulus (G) of the amorphous phase. The first option was by assuming the chain bundle to be a cylinder, and tensile forces to be applied on it. The maximum force that the cylinder can withstand without failure is the peak force, 6840 pN for 100 m/s pulling velocity. The cross-section area of the cylinder was 10.12 nm^2, with a radius of 3.18 nm. The strain in the cylinder at the peak load was 0.141 by considering the shift in the center-of-mass along the pulling direction. Therefore, the Young's modulus of the cylindrical chain bundle was E = peak force/(cross-section area $\times$ strain) = 4.79 GPa, $G = E/(2 \times (1 + \nu))$ = 1.66 GPa, where ν is the Poisson's ratio. Thus, the shear modulus for our viscoelastic amorphous phase was 1.66 GPa.

As the second option, we estimated the shear modulus from the ratio of shear stress and strain, where shear stress is the force per unit area for the relative sliding of the peptide chains. Fig. S3 shows the shear stress-strain behavior for the 8-chain bundle pulled at 10 m/s. We considered the initial slope of the shear stress-strain curve and obtained a G of $\sim$1.7 GPa in agreement with the other estimate above.

Supporting figure legends

Supporting Figure S1. Total friction force per residue (F/N) as a function of pulling velocity (V) for three simulation systems, a 4-chain, an 8-chain and a 24-chain bundle. There is no significant difference in the F/N for the 8-chain and 24-chain bundle.

Supporting Figure S2. A shear stress-strain curve for the 8-chain bundle from the amorphous phase of spider silk for 10 m/s pulling velocity with error bar (gray). In the initial portion of the curve (till 0.07), the shear stress varies linearly with the shear strain. The linear dotted line shows the slope of the initial portion of the curve.

Supporting Figure S3. General behavior of the amorphous phase under constant strain and stress loading from FEM. (A) A rectangular box of the amorphous material loaded to a constant strain of 0.1. (B) The developed stress relaxes from 75 MPa to negligible values within a few ms. (C) In a creep test, a constant stress of 0.14 MPa was applied to the material, (D) resulting in an instantaneous elastic straining, followed by a creep in strain up to the equilibrium value of 4.5%.

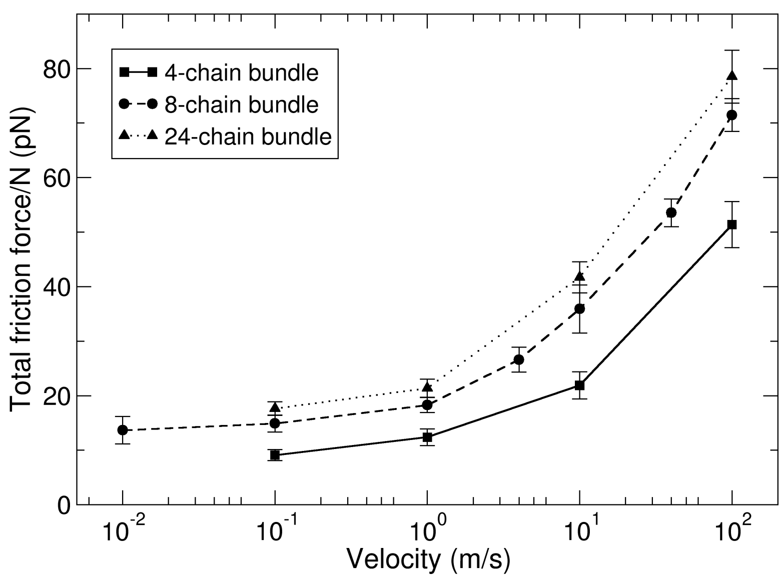
4-chain bundle
8-chain bundle
24-chain bundle
Total friction force/N (pN)
0
20
40
60
80
10^{-2}
10^{-1}
10^{0}
10^{1}
10^{2}
Velocity (m/s)

Figure S1.

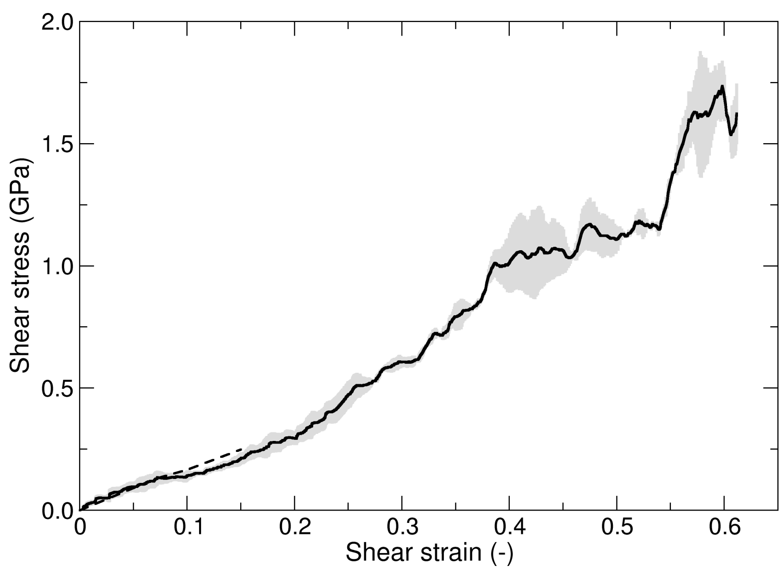

Figure S2.

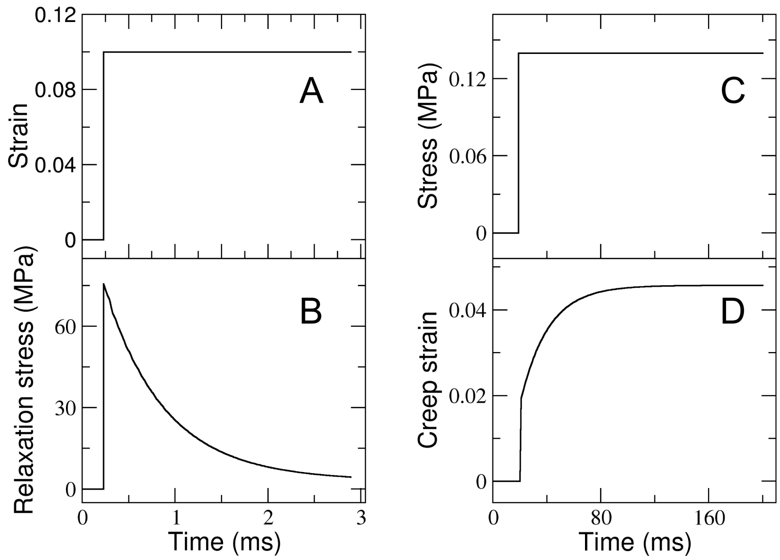
A
B
C
D
Strain
Relaxation stress (MPa)
Stress (MPa)
Creep strain
Time (ms)
Time (ms)
0.12
0.08
0.04
0
60
30
0
0.12
0.06
0
0.04
0.02
0
0
1
2
3
0
80
160

Figure S3.