Designing high performance polymer nanocomposites by incorporating robustness-controlled polymeric nanoparticles: insights from molecular dynamics

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Details of simulation model

In this study, both η = 0 and η = 1.0 were obtained same mean-square radius gyration, which was equal to approximately 5σ. The polymer matrix chains were fixed to be 100 beads, and each system contained 300 chains. For star-shaped nanoparticle (SSPNs), there are two sections: First section changed the hardness of NP, hence the η of star-shape systems were equal to 0.2, 0.4, 0.6, 0.8, respectively, and the arm length, *L*, was set as 10σ (Figure S1); Second section changed the arm length of star-shaped NP, hence *L* was equal to 5σ, 10σ, 15σ, 20σ, respectively, and η was set as 0.8. For all systems, the mass fraction of NPs was fixed as 20%. (Figure S2)

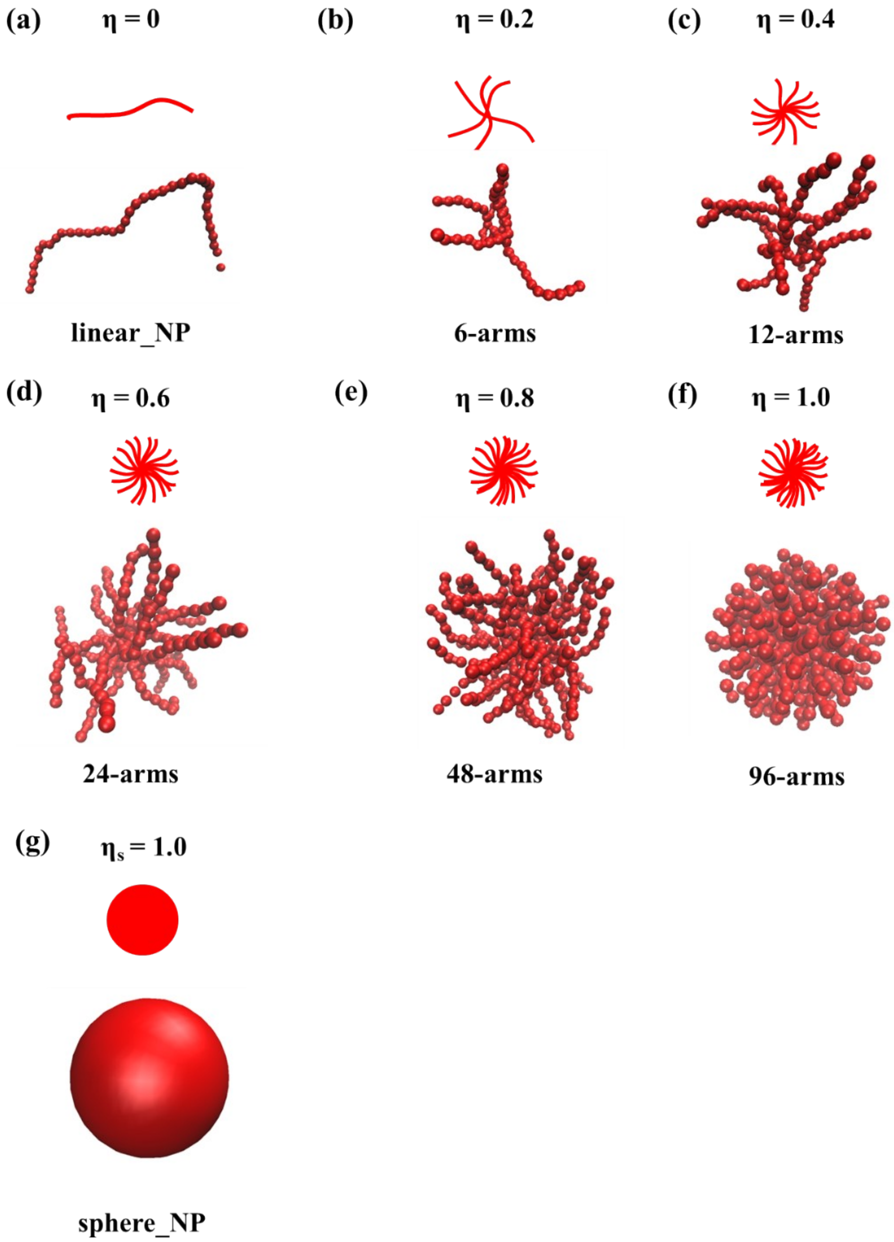


Figure S1. The schematic diagram of NP, including (a) $\eta = 0$; (b) $\eta = 0.2$; (c) $\eta = 0.4$; (d) $\eta = 0.6$; (e) $\eta = 0.8$; (f) $\eta = 1.0$; (g) $\eta_s = 1.0$; Noted that both linear NP and SSPNs were composed by the red-sphere bead that diameter was equal to 1σ.

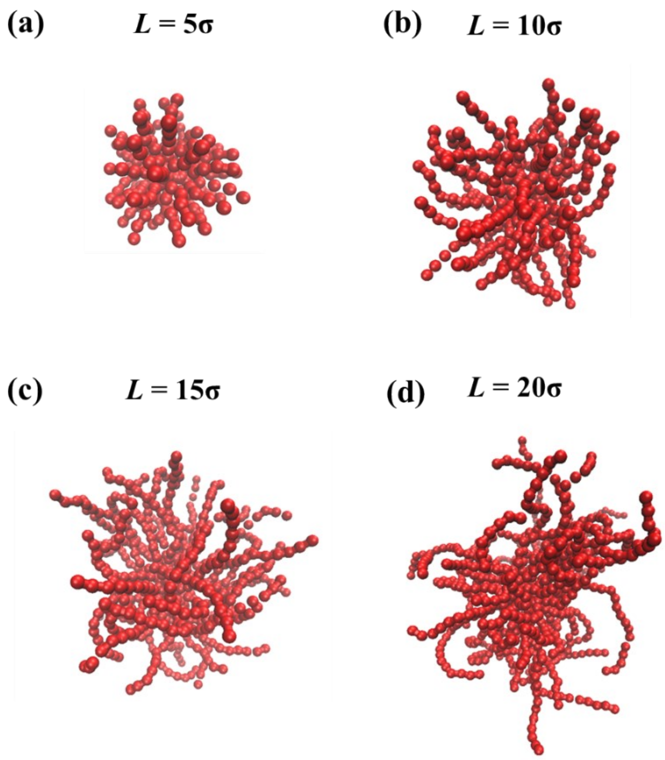


Figure S2. The schematic diagram for SSPNs with different L, where (a) L =5σ; (b) L =10σ; (c) L =15σ; (d) L =20σ. Noted that η was set as 0.8.

Table S1. the mean-root square radius of gyration (R_g) of NPs with different η.

system	R_g (σ)
$\eta = 0$	5.1108
$\eta = 0.2$	4.829
$\eta = 0.4$	4.94
$\eta = 0.6$	4.923
$\eta = 0.8$	4.78842
$\eta = 1.0$	4.9951
$\eta_s = 1.0$	5

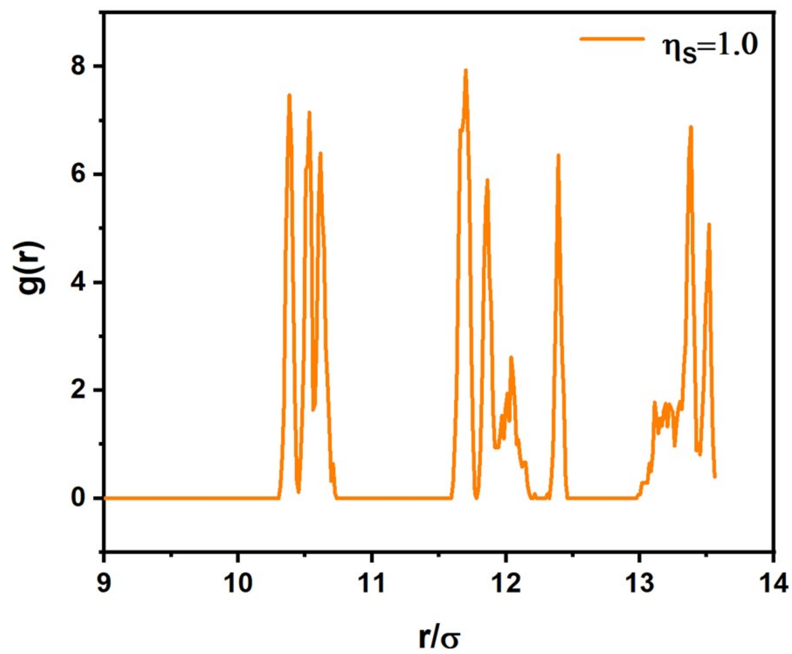


Figure S3. The radical distribution function of NP when η_s = 1.0.

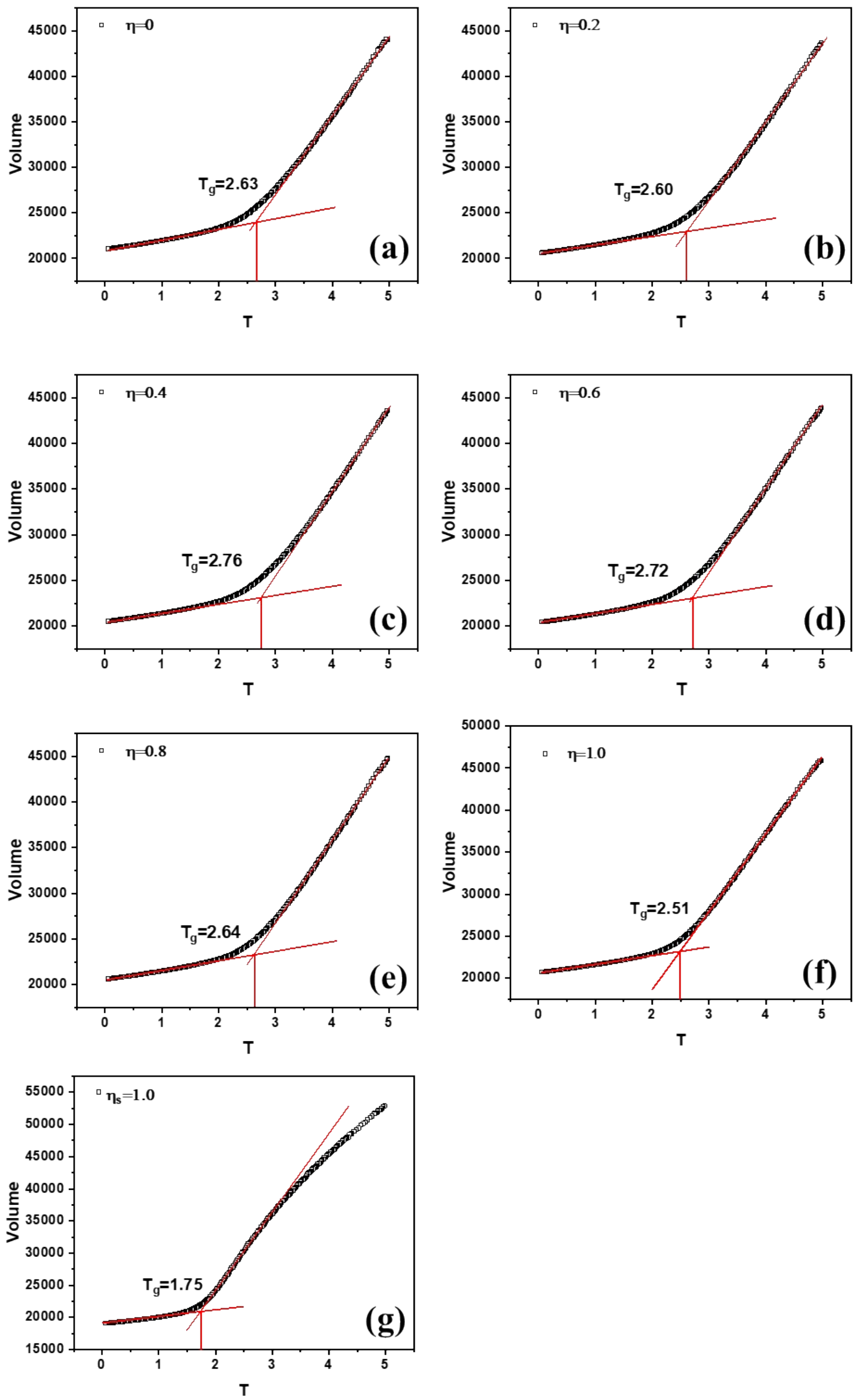
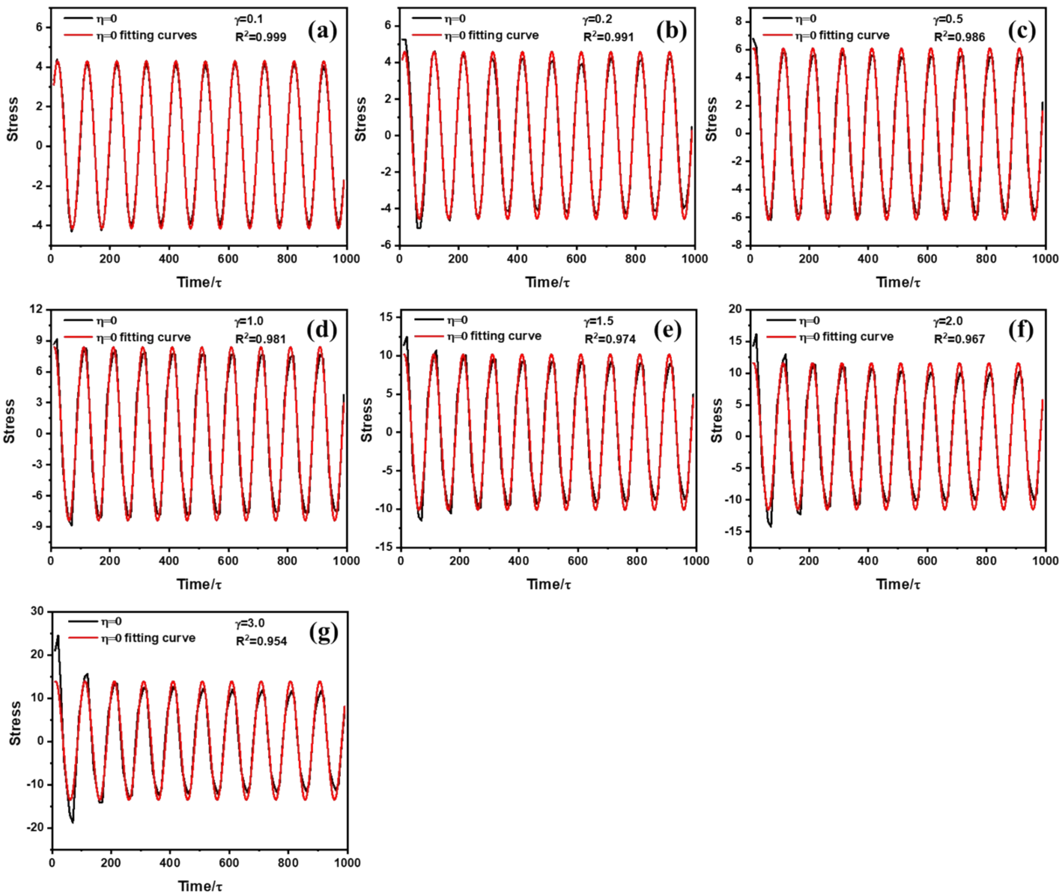


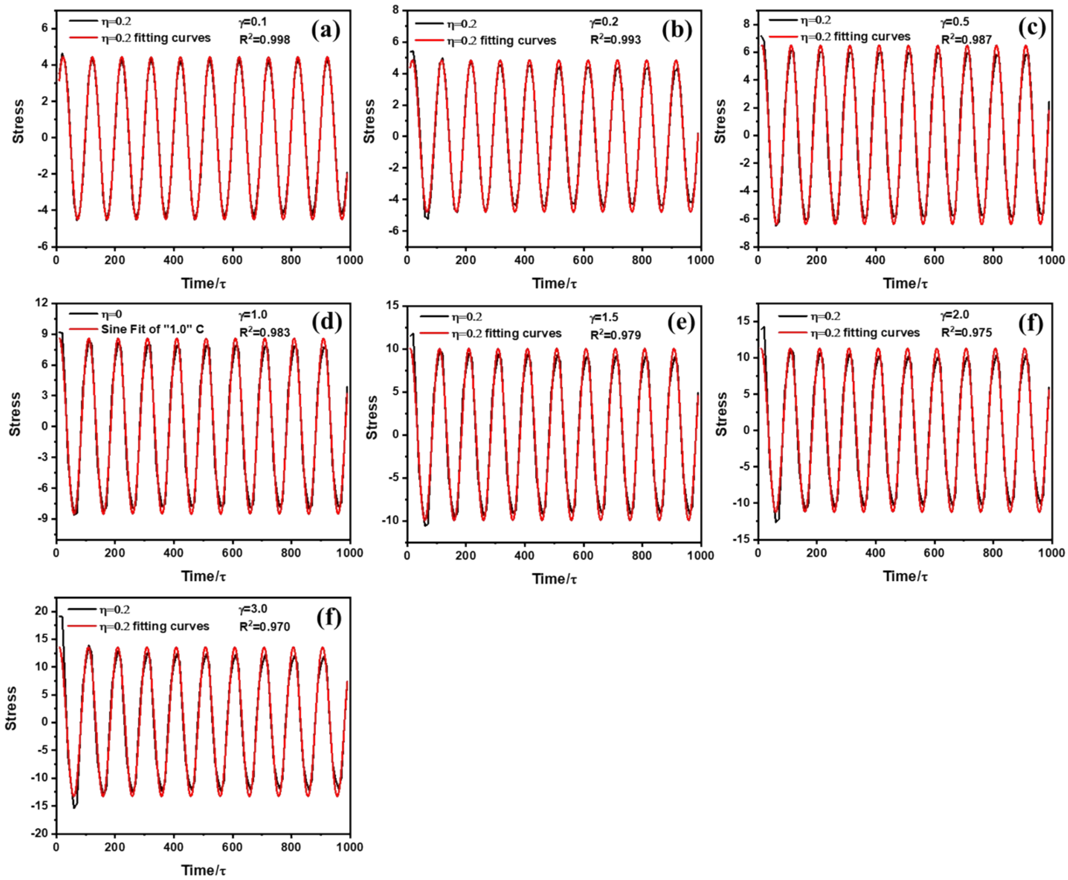
Figure S4. The glass transition temperature for PNCs when: (a) $\eta = 0$; (b) $\eta = 0.2$; (c) $\eta = 0.4$; (d) $\eta = 0.6$; (e) $\eta = 0.8$; (f) $\eta = 1.0$; (g) $\eta_s = 1.0$

η=0

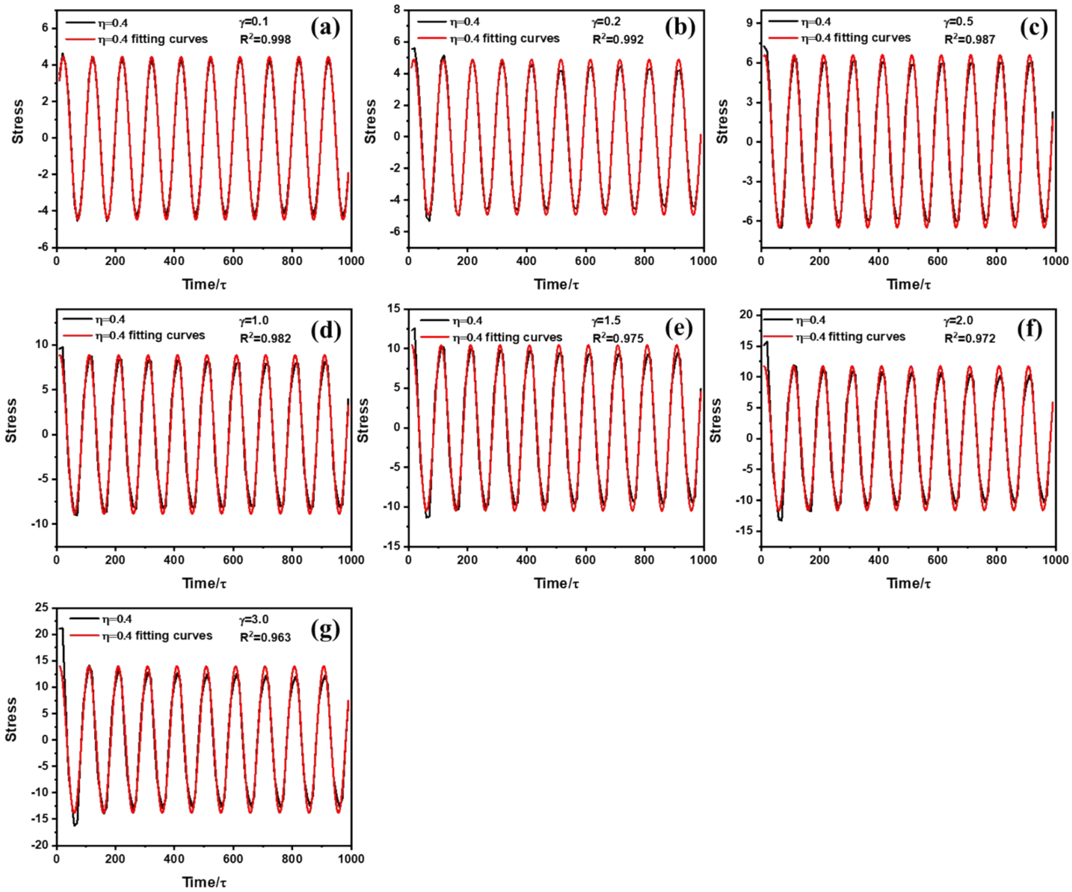


η=0
η=0 fitting curves
γ=0.1
R²=0.999
(a)
η=0
η=0 fitting curve
γ=0.2
R²=0.991
(b)
η=0
η=0 fitting curve
γ=0.5
R²=0.986
(c)
η=0
η=0 fitting curve
γ=1.0
R²=0.981
(d)
η=0
η=0 fitting curve
γ=1.5
R²=0.974
(e)
η=0
η=0 fitting curve
γ=2.0
R²=0.967
(f)
η=0
η=0 fitting curve
γ=3.0
R²=0.954
(g)
Stress
Time/τ

η=0.2

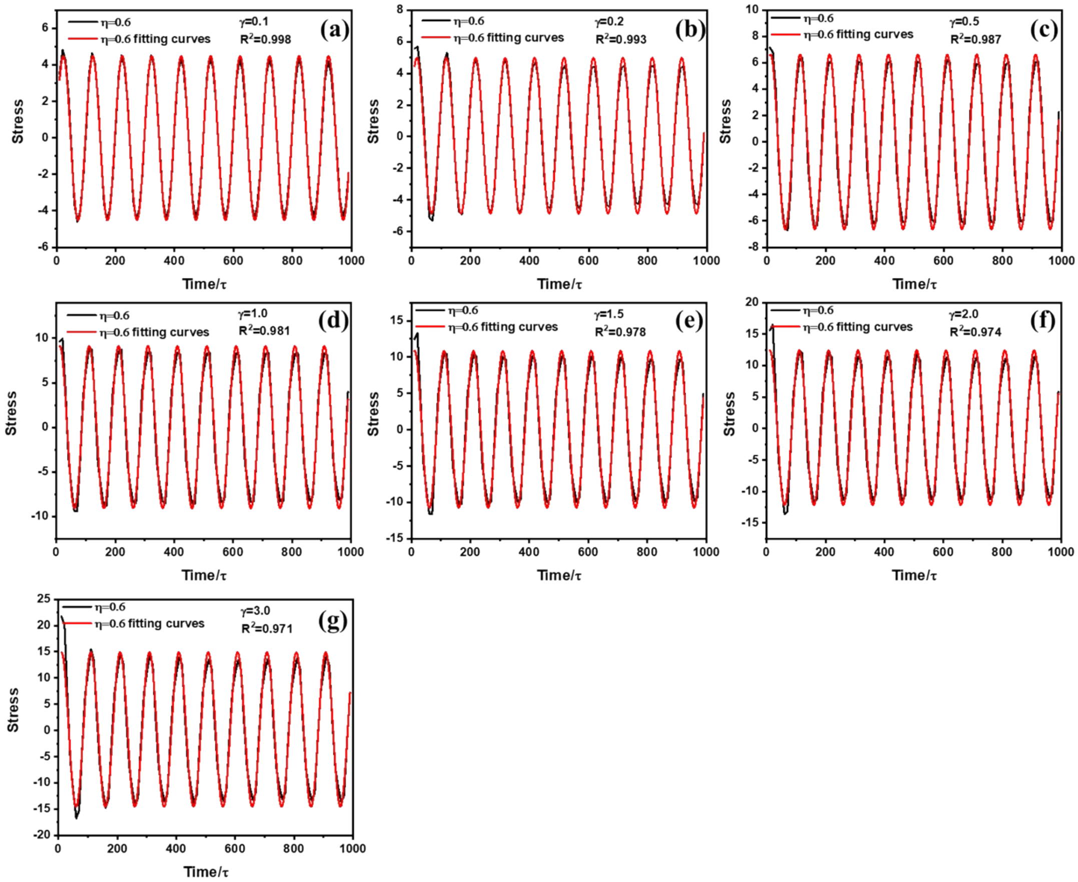

η=0.2
η=0.2 fitting curves
γ=0.1
R²=0.998
(a)
η=0.2
η=0.2 fitting curves
γ=0.2
R²=0.993
(b)
η=0.2
η=0.2 fitting curves
γ=0.5
R²=0.987
(c)
η=0
Sine Fit of "1.0" C
γ=1.0
R²=0.983
(d)
η=0.2
η=0.2 fitting curves
γ=1.5
R²=0.979
(e)
η=0.2
η=0.2 fitting curves
γ=2.0
R²=0.975
(f)
η=0.2
η=0.2 fitting curves
γ=3.0
R²=0.970
(f)
Stress
Time/τ

η=0.4

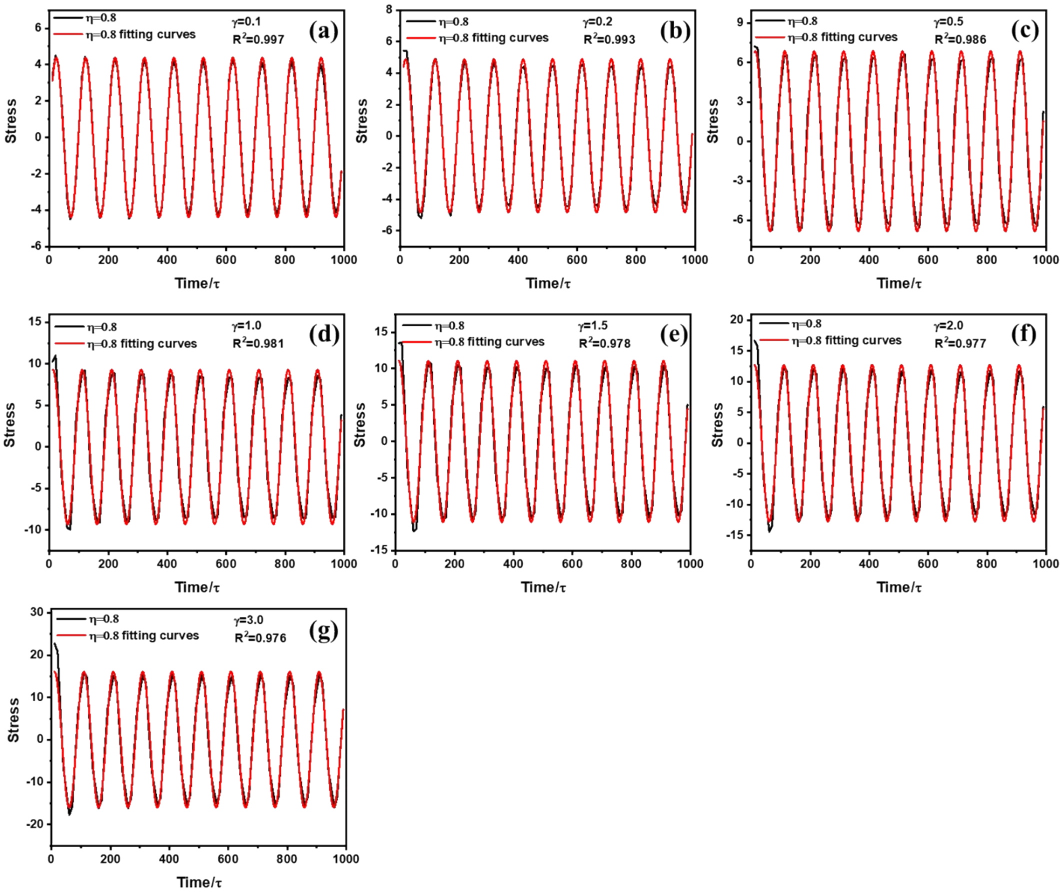


(a)
η=0.4
η=0.4 fitting curves
γ=0.1
R²=0.998
(b)
η=0.4
η=0.4 fitting curves
γ=0.2
R²=0.992
(c)
η=0.4
η=0.4 fitting curves
γ=0.5
R²=0.987
(d)
η=0.4
η=0.4 fitting curves
γ=1.0
R²=0.982
(e)
η=0.4
η=0.4 fitting curves
γ=1.5
R²=0.975
(f)
η=0.4
η=0.4 fitting curves
γ=2.0
R²=0.972
(g)
η=0.4
η=0.4 fitting curves
γ=3.0
R²=0.963
Stress
Time/τ

η=0.6

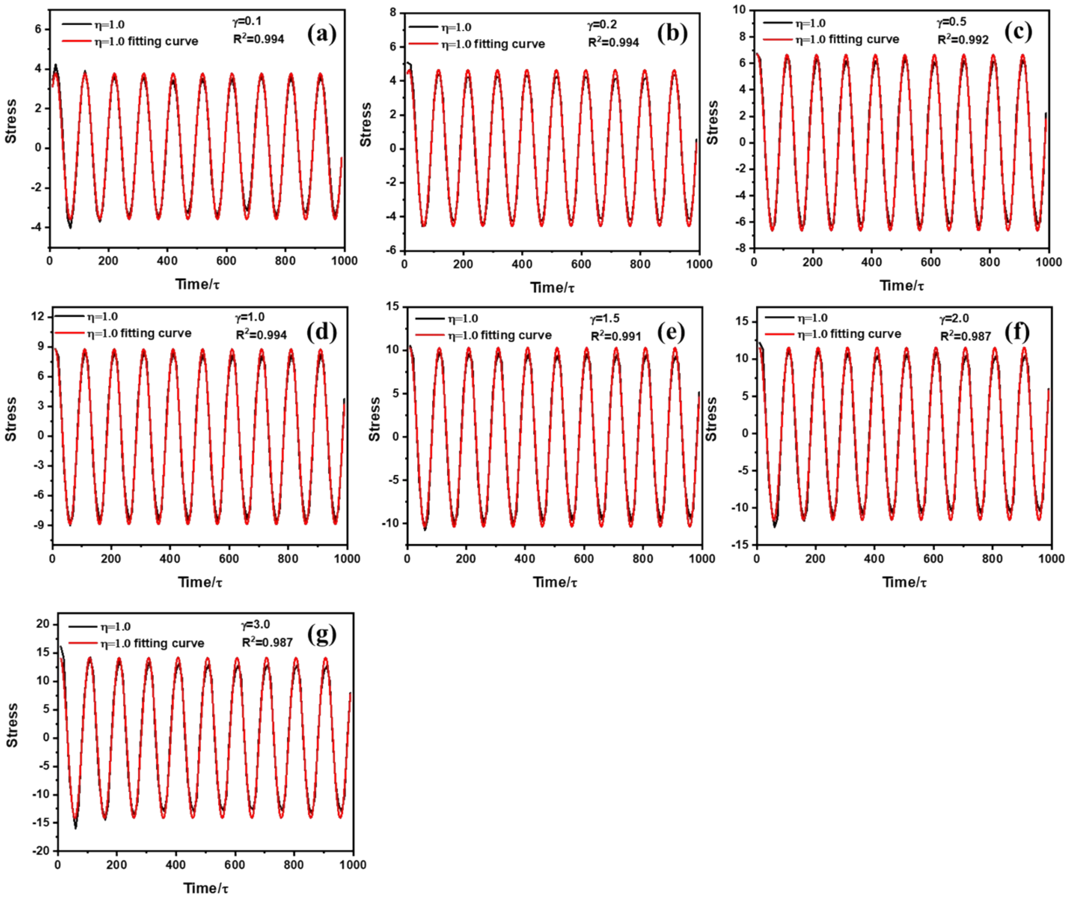

η=0.6
η=0.6 fitting curves
γ=0.1
R²=0.998
(a)
γ=0.2
R²=0.993
(b)
γ=0.5
R²=0.987
(c)
γ=1.0
R²=0.981
(d)
γ=1.5
R²=0.978
(e)
γ=2.0
R²=0.974
(f)
γ=3.0
R²=0.971
(g)
Stress
Time/τ

η=0.8



(a)
η=0.8
η=0.8 fitting curves
γ=0.1
R^2=0.997
(b)
γ=0.2
R^2=0.993
(c)
γ=0.5
R^2=0.986
(d)
γ=1.0
R^2=0.981
(e)
γ=1.5
R^2=0.978
(f)
γ=2.0
R^2=0.977
(g)
γ=3.0
R^2=0.976
Stress
Time/τ

η=1.0



η=1.0
η=1.0 fitting curve
γ=0.1
R²=0.994
(a)
γ=0.2
R²=0.994
(b)
γ=0.5
R²=0.992
(c)
γ=1.0
R²=0.994
(d)
γ=1.5
R²=0.991
(e)
γ=2.0
R²=0.987
(f)
γ=3.0
R²=0.987
(g)
Stress
Time/τ

η_s=1.0

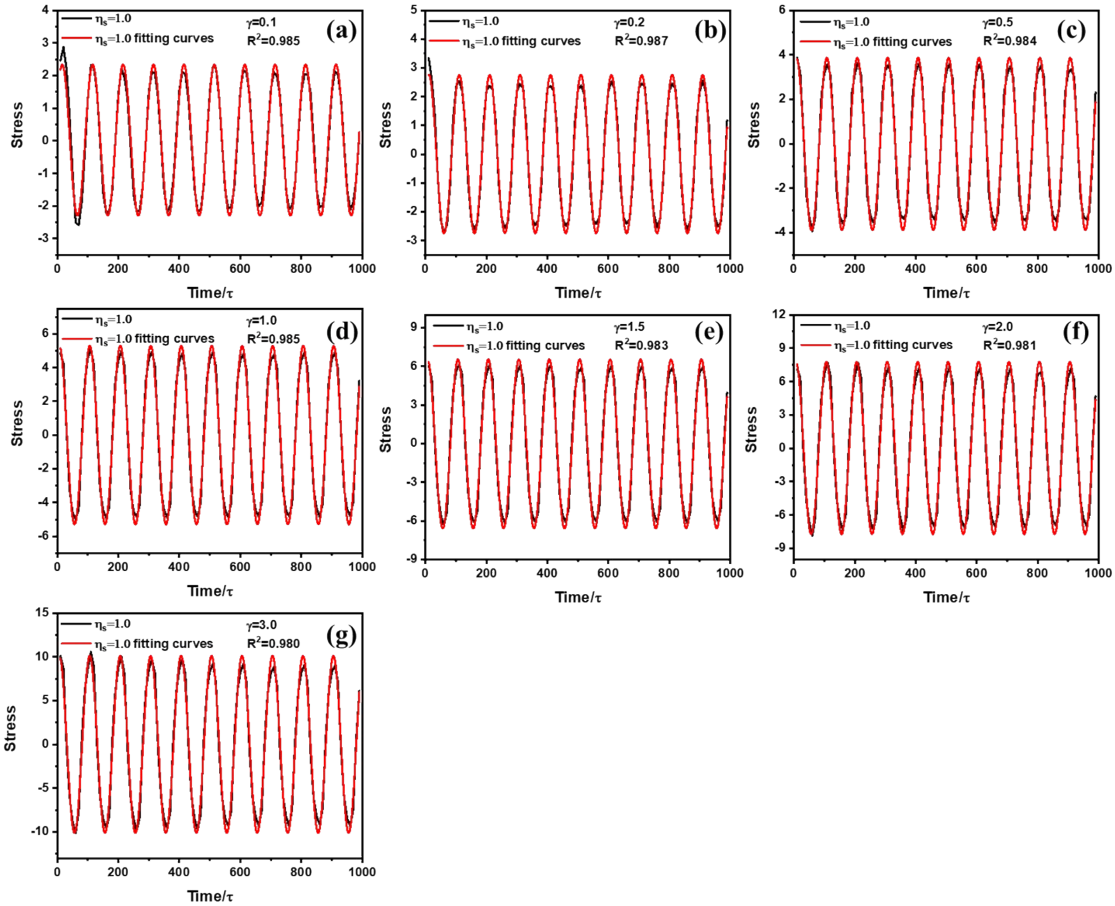


Figure S5. Shear stress-strain curves of SSPNs with different η, where (a) γ=0.1; (b) γ=0.2; (c) γ=0.5; (d) γ=1.0; (e) γ=1.5; (f) γ=2.0; (g) γ=3.0;

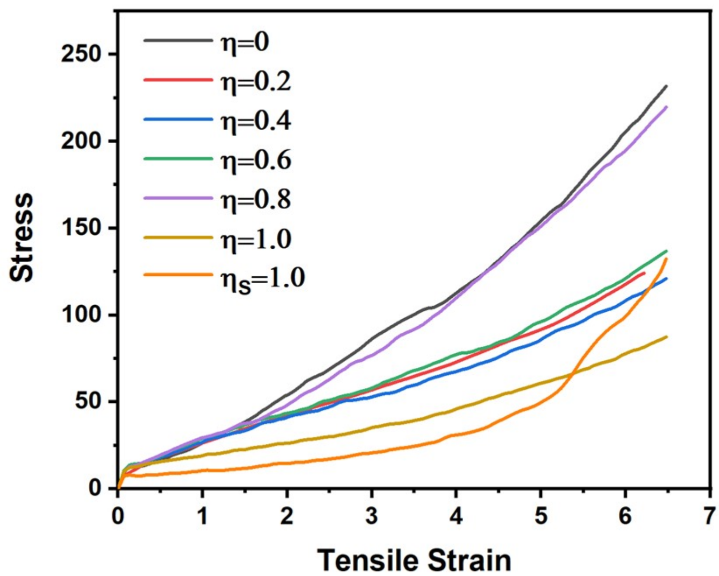


Figure S6. The stress-strain curve for different η.

Table S2. R_g of various L. Noted that $\eta = 0.8$

L (σ)	R_g (σ)
5	3.66
10	4.79
15	8.85
20	9.07

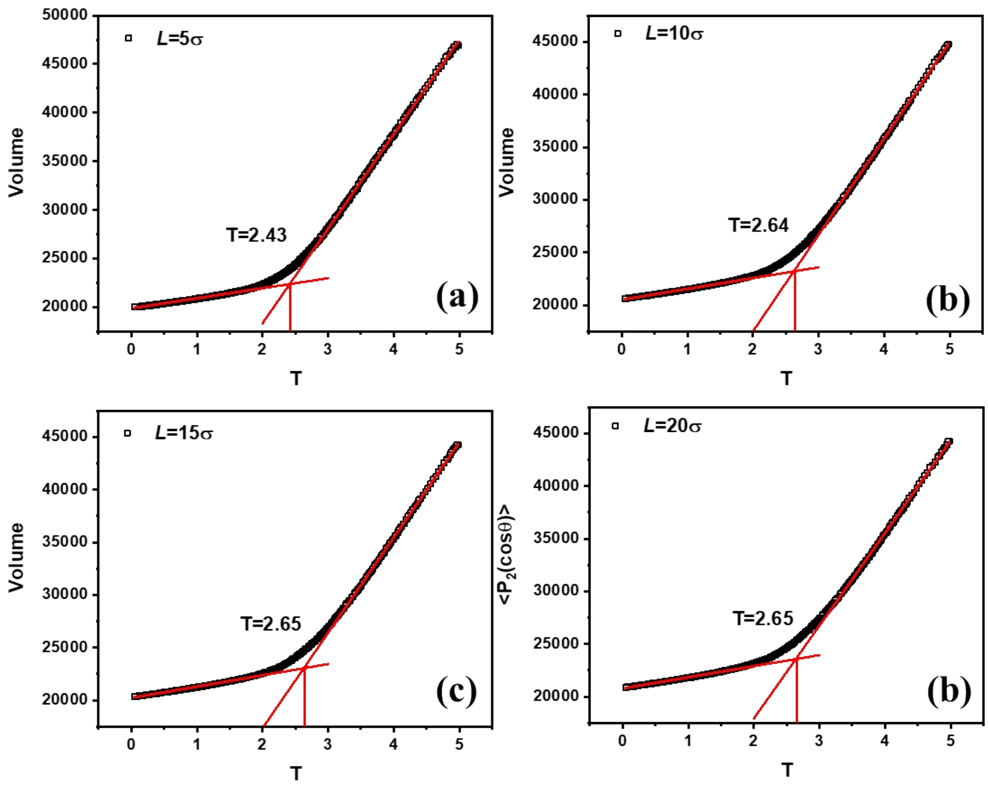


Figure S7. The glass transition temperature for PNCs when :(a) $L = 5\sigma$; (b) $L = 10\sigma$; (c) $L = 15\sigma$; (d) $L = 20\sigma$. Noted that $\eta = 0.8$.

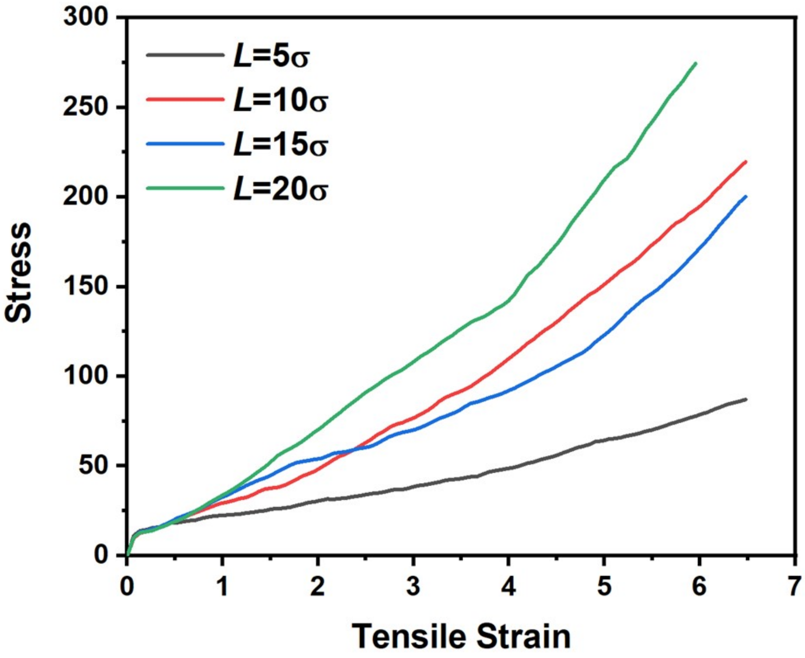
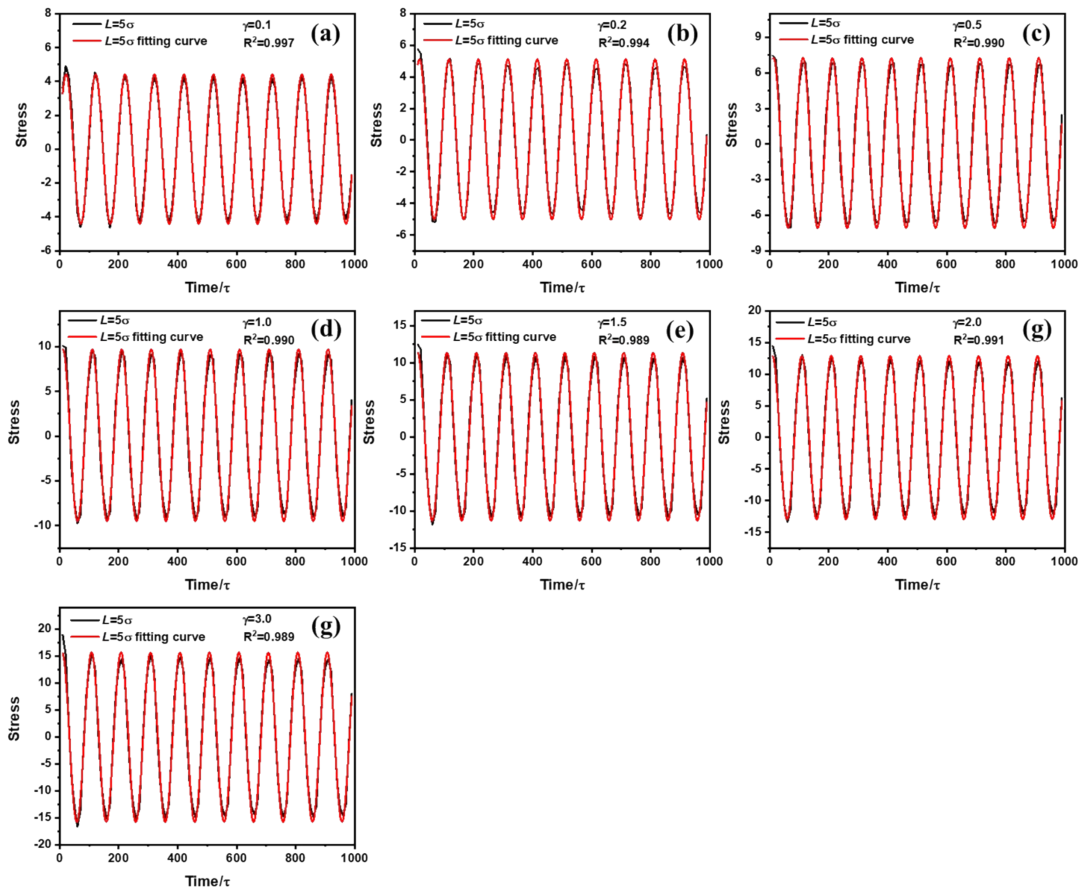
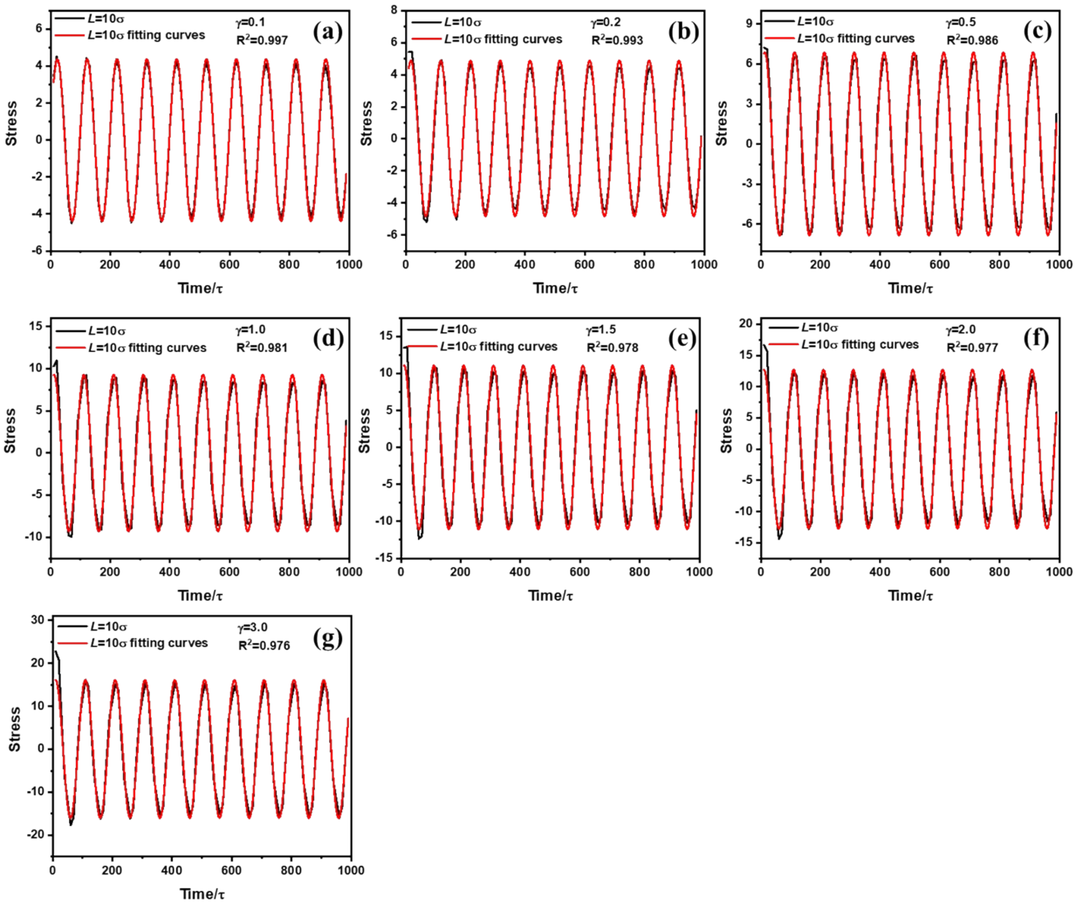


Figure S8. The stress-strain curve for different η.

***L* = 5σ**

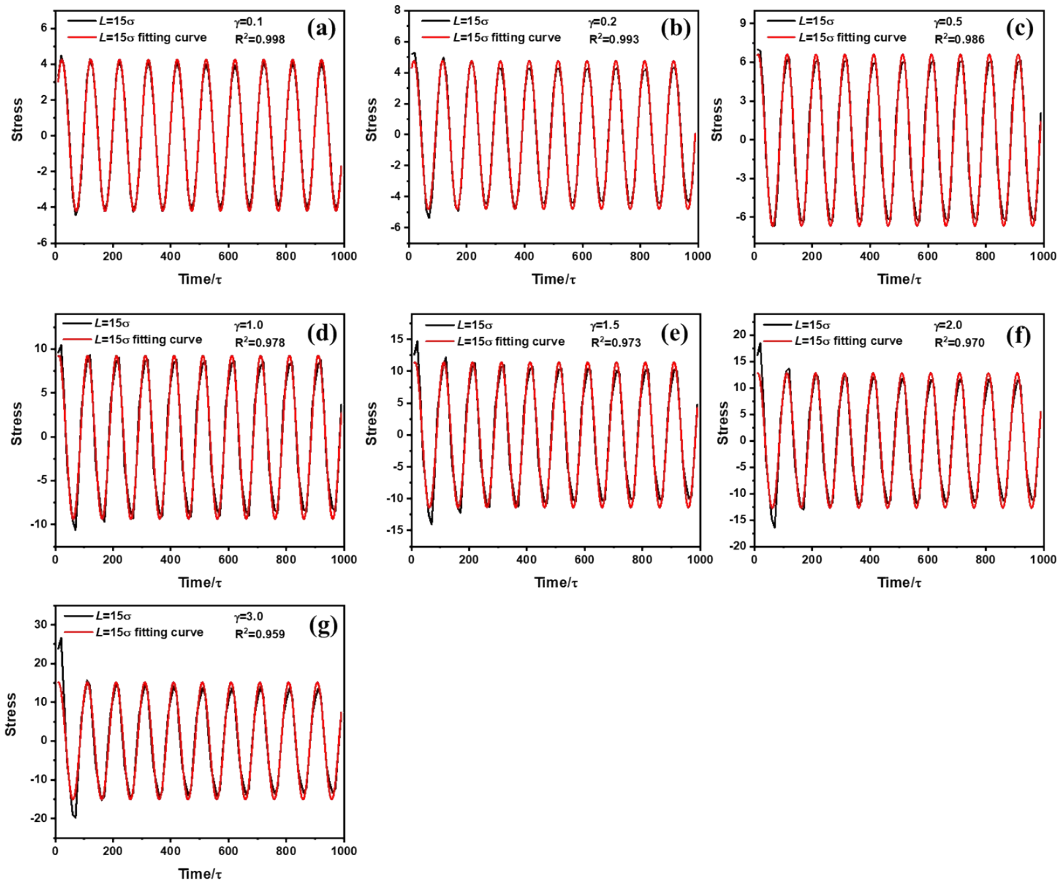


***L* = 10σ**



L=10σ
L=10σ fitting curves
γ=0.1
R²=0.997
(a)
γ=0.2
R²=0.993
(b)
γ=0.5
R²=0.986
(c)
γ=1.0
R²=0.981
(d)
γ=1.5
R²=0.978
(e)
γ=2.0
R²=0.977
(f)
γ=3.0
R²=0.976
(g)
Stress
Time/τ

***L* = 15σ**



(a)
L=15σ
L=15σ fitting curve
γ=0.1
R²=0.998
(b)
L=15σ
L=15σ fitting curve
γ=0.2
R²=0.993
(c)
L=15σ
L=15σ fitting curve
γ=0.5
R²=0.986
(d)
L=15σ
L=15σ fitting curve
γ=1.0
R²=0.978
(e)
L=15σ
L=15σ fitting curve
γ=1.5
R²=0.973
(f)
L=15σ
L=15σ fitting curve
γ=2.0
R²=0.970
(g)
L=15σ
L=15σ fitting curve
γ=3.0
R²=0.959
Stress
Time/τ

***L* = 20σ**

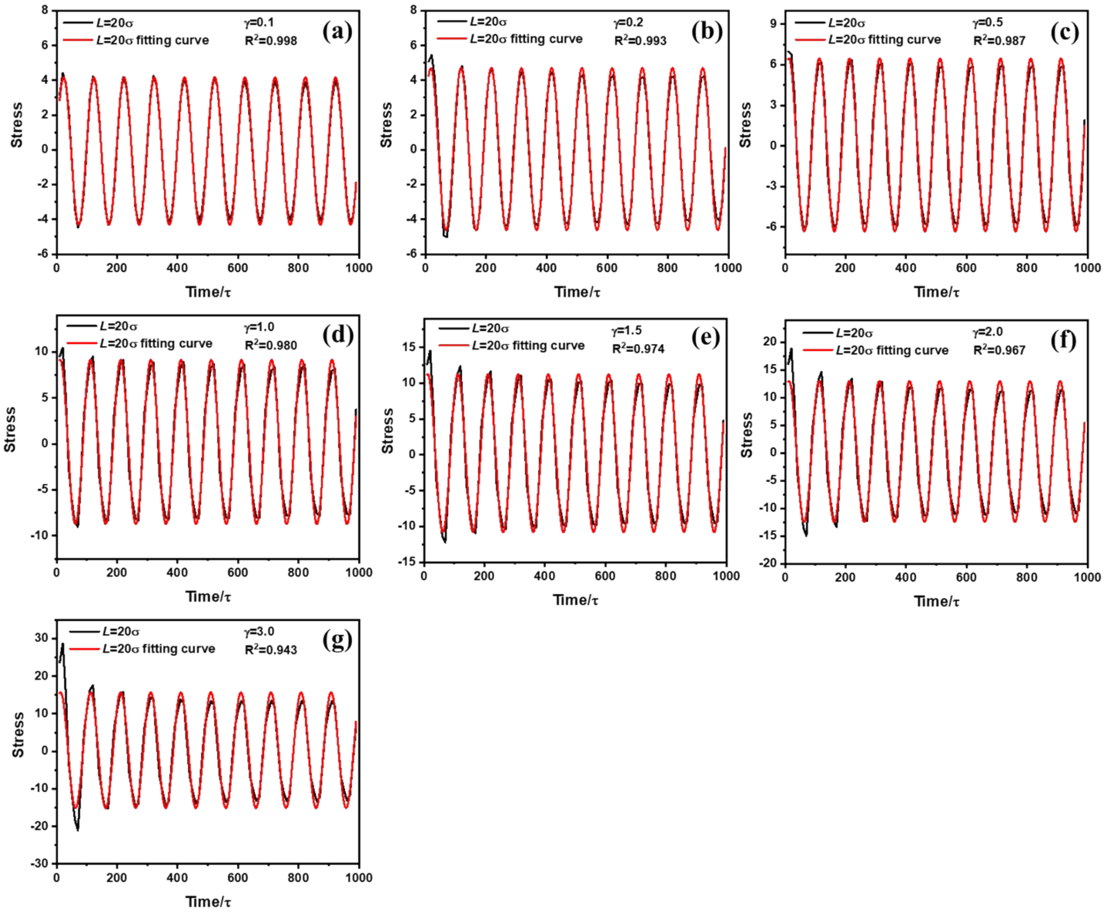


Figure S9. Shear stress-strain curves of SSPNs with different L, where (a) γ=0.1; (b) γ=0.2; (c) γ=0.5; (d) γ=1.0; (e) γ=1.5; (f) γ=2.0; (g) γ=3.0;