Electronic Supplementary Information for

Flow-induced structure and properties of multiwall carbon nanotube/polydimethylsiloxane composites

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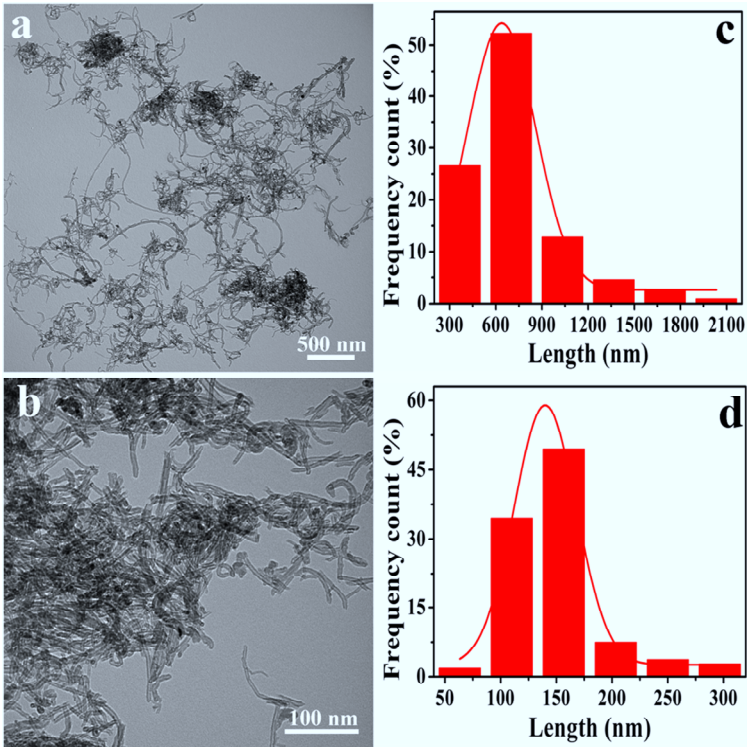

Fig. S1 TEM images and length distributions of MWNT2 (a and c) and MWNT6 (b and d).

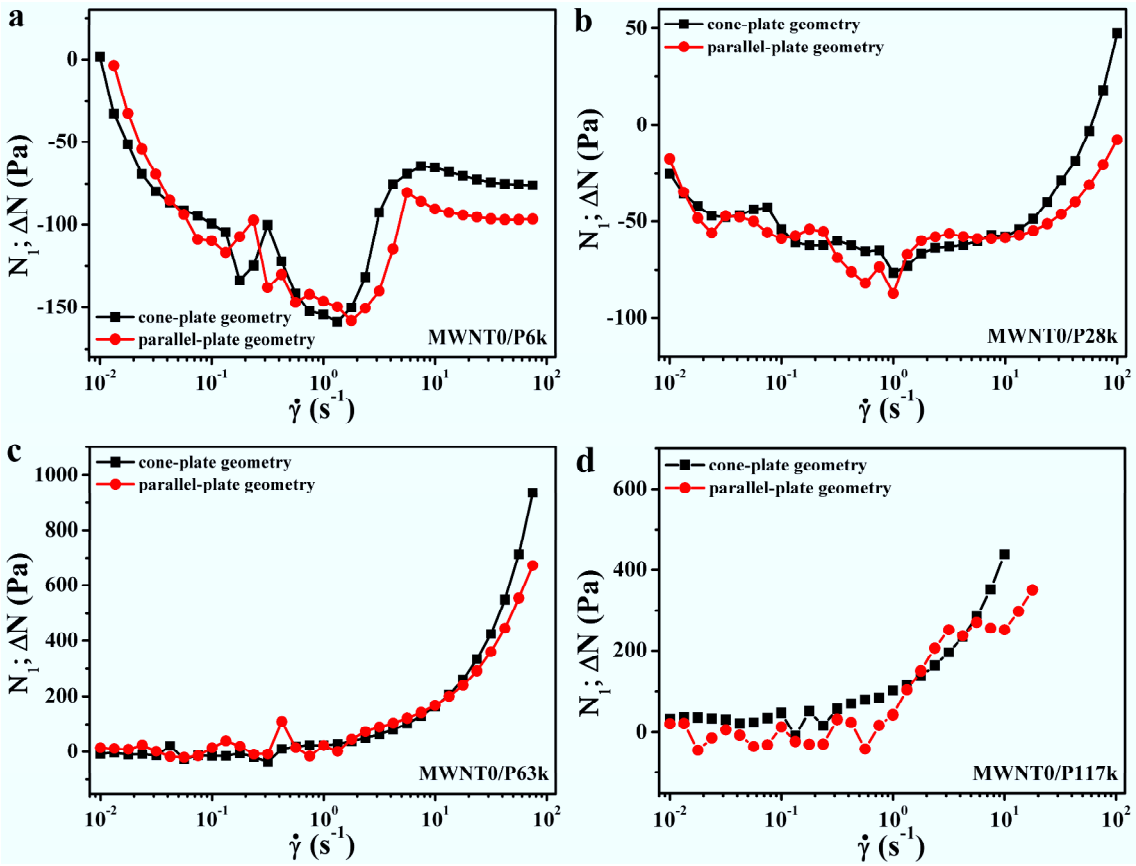

Fig. S2 Normal stress difference (N_1 and ΔN) versus shear rate measured by cone-plate and parallel-plate geometries for 2 wt % MWNT0/PDMS composites.

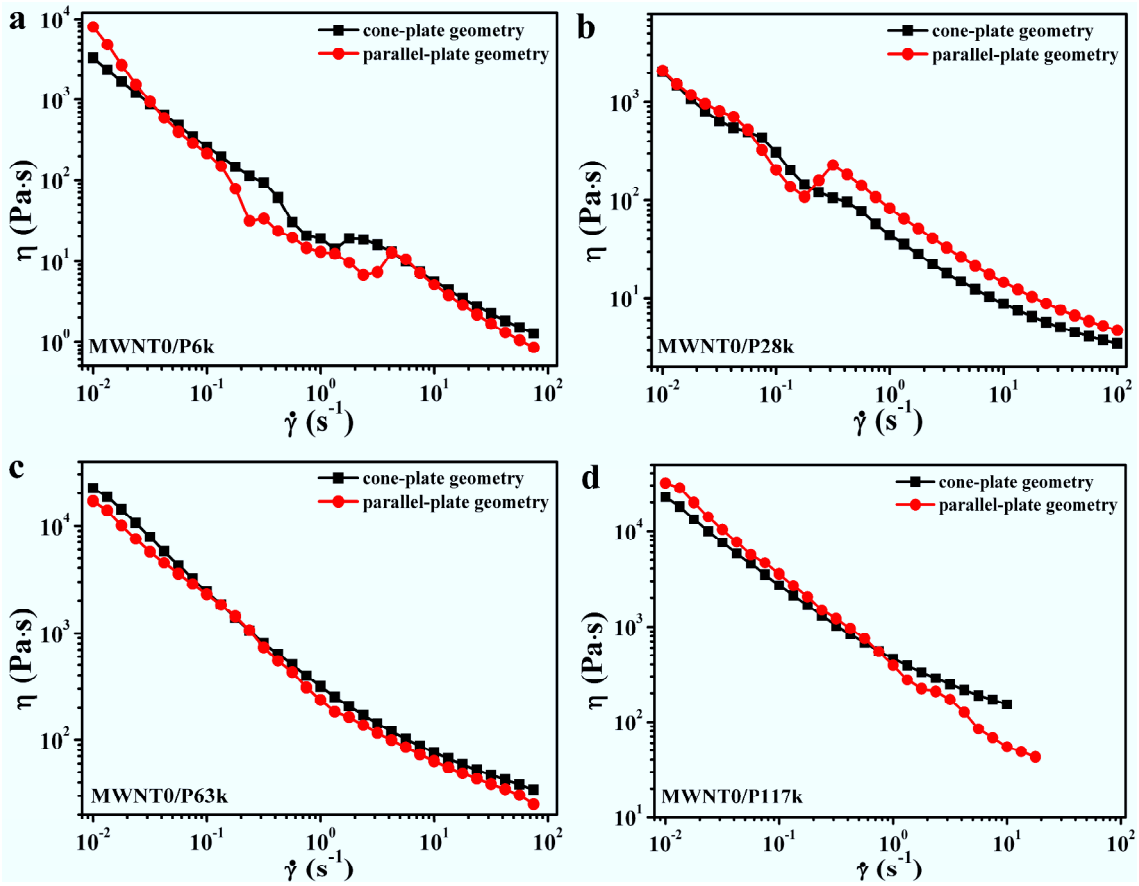

Fig. S3 Viscosity versus shear rate measured by cone-plate and parallel-plate geometries for 2 wt % MWNT0/PDMS composites.

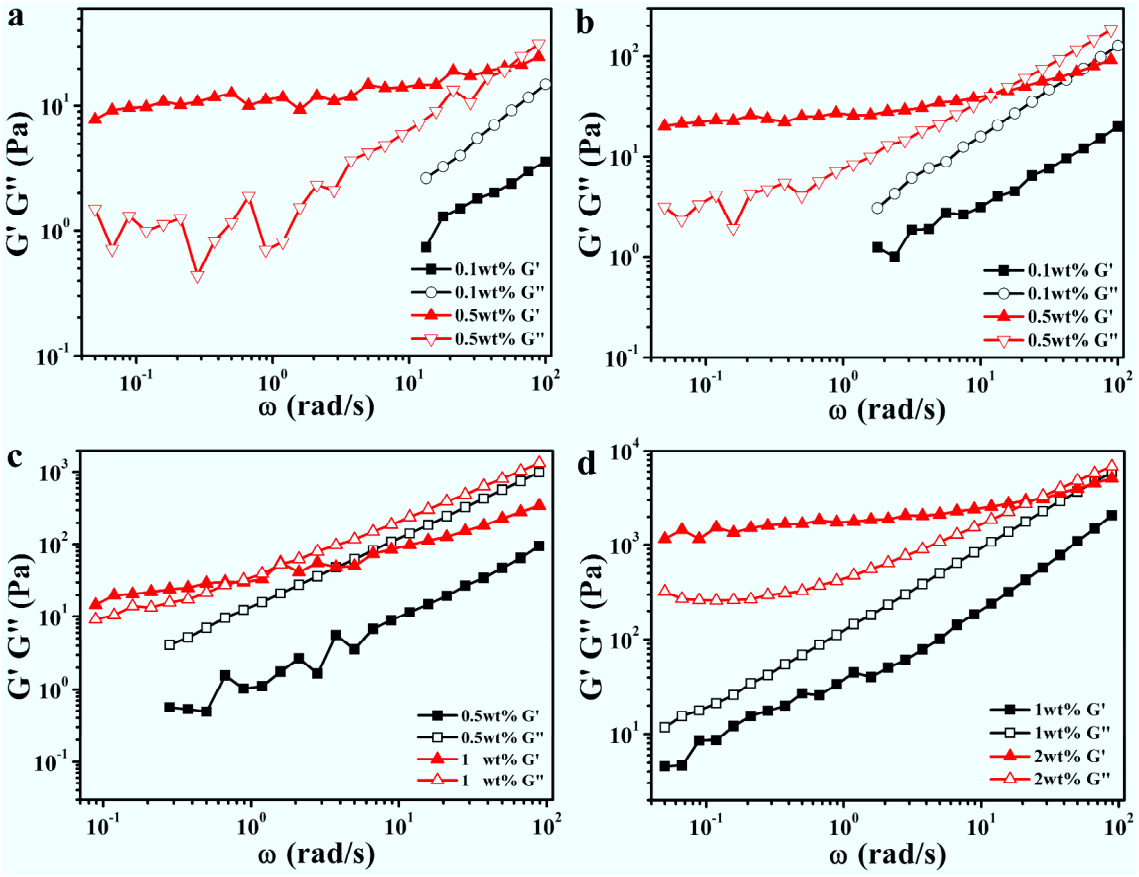

Fig. S4 Frequency sweep results of MWNT0/P6k (a), MWNT0/P28k (b), MWNT0/P63k (c) and MWNT0/P117k composites (d).

For 0.5 wt % MWNT0/P6k and 0.5 wt % MWNT0/P28k, the storage modulus (*G*') is larger than loss modulus (*G*''), indicating the formation of network structure.[S1–S3] However, for 0.1 wt % MWNT0/P6k and 0.1 wt % MWNT0/P28k, the *G*' and *G*'' at low frequency cannot be accurately measured as the torque is close to the resolution limit. At high frequency, *G*'' is larger than *G*', suggesting that no network structure forms.[S1–S3]

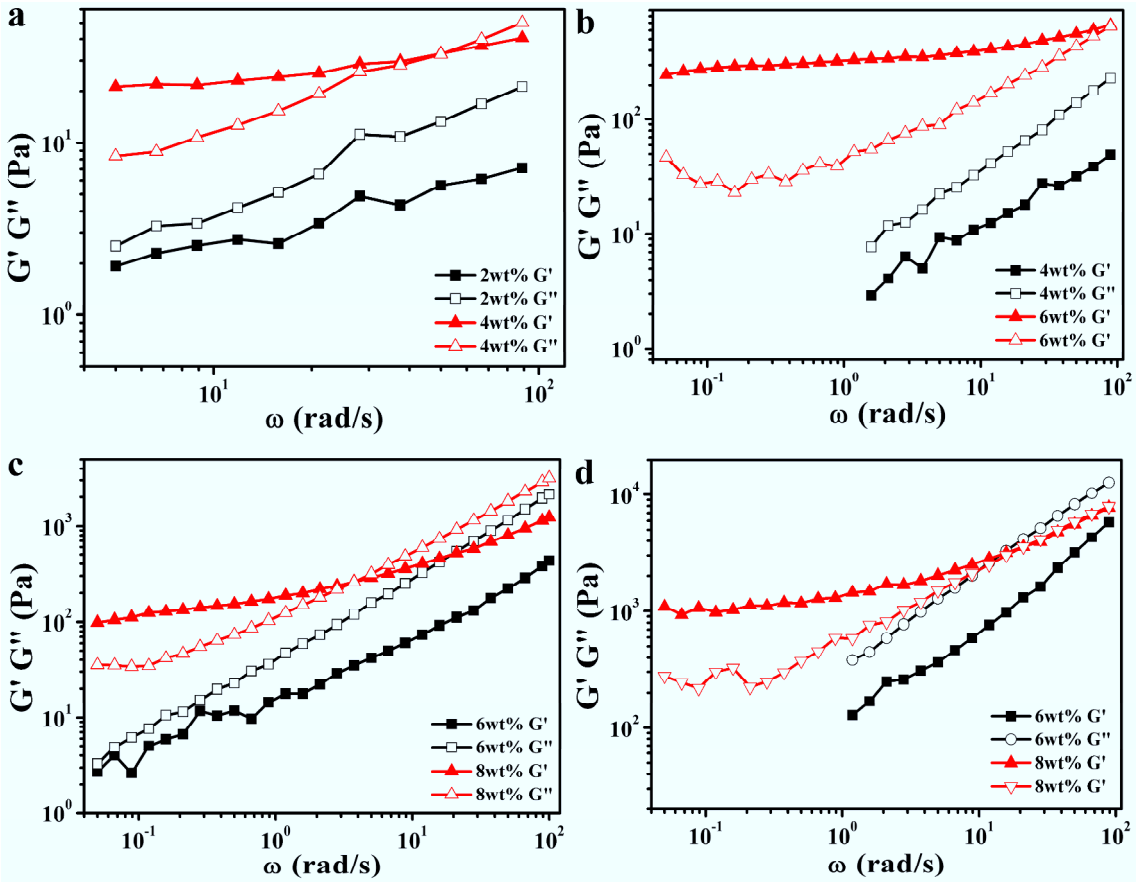

Fig. S5 Frequency sweep results of MWNT2/P6k (a), MWNT2/P28k (b), MWNT2/P63k (c) and MWNT2/P117k composites (d).

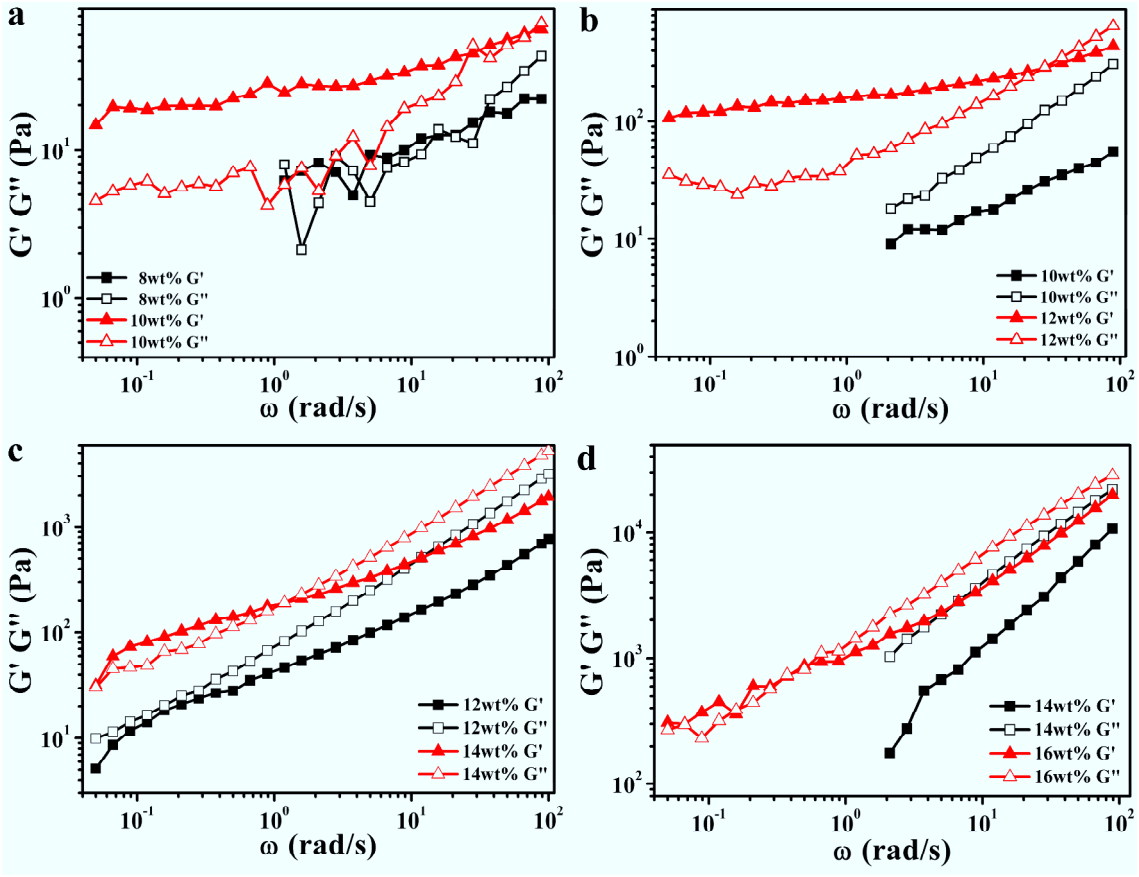

Fig. S6 Frequency sweep results of MWNT6/P6k (a), MWNT6/P28k (b), MWNT6/P63k (c) and MWNT6/P117k composites (d).

Influence of PDMS molecular weight on the rheological properties of MWNT/PDMS composites

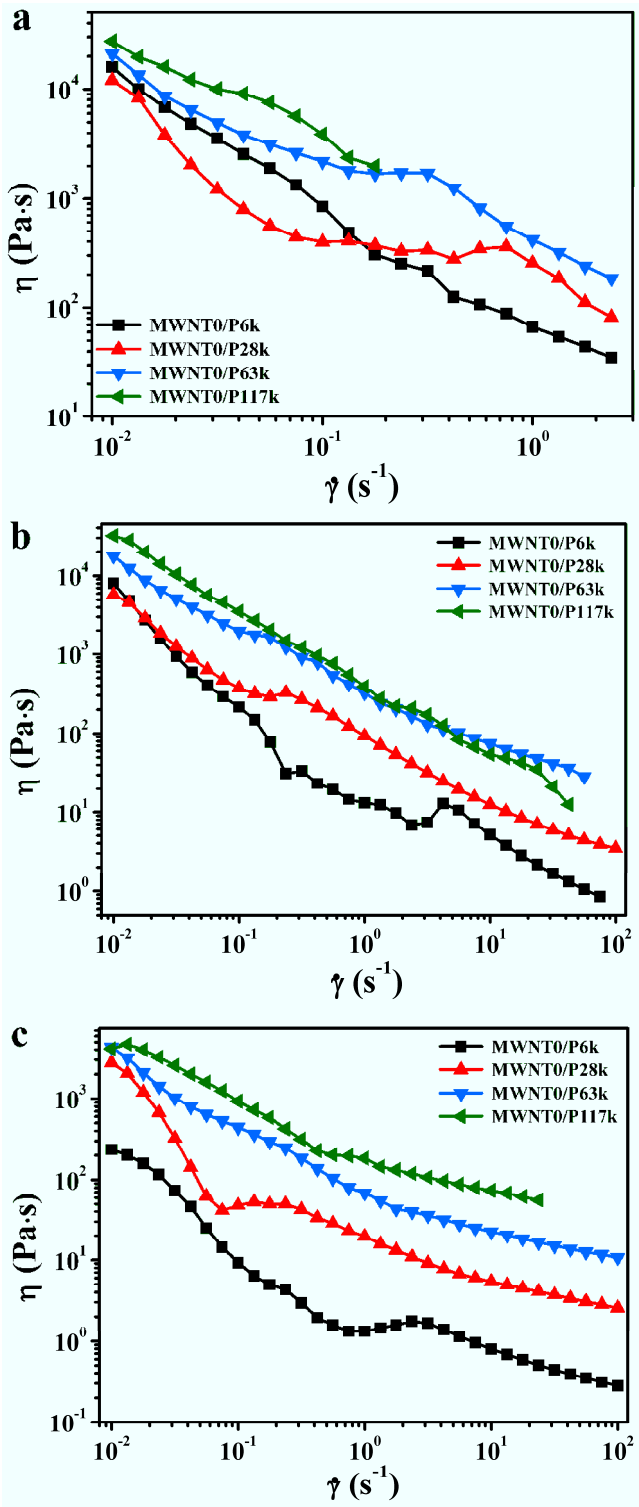

Fig. S7 Viscosity (η) versus shear rate ($\dot{\gamma}$) for MWNT0/PDMS composites with 4 wt % MWNT0 (a), 2 wt % MWNT0 (b) and 1 wt % MWNT0 (c). The gap used in the rheological experiments is about 0.9 mm.

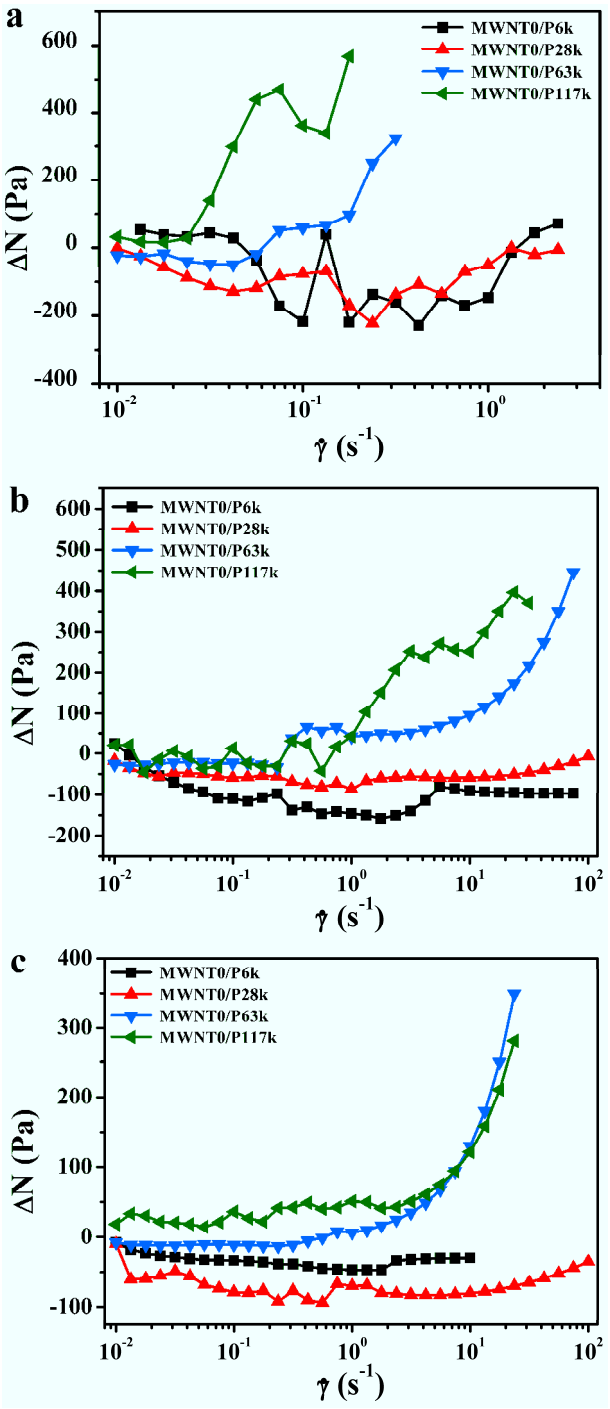

Fig. S8 Normal stress differences (ΔN) versus shear rate ($\dot{\gamma}$) for MWNT0/PDMS composites with 4 wt % MWNT0 (a), 2 wt % MWNT0 (b) and 1 wt % MWNT0 (c). The gap used in the rheological experiments is about 0.9 mm.

Influence of MWNT aspect ratio on the rheological properties of MWNT/PDMS composites

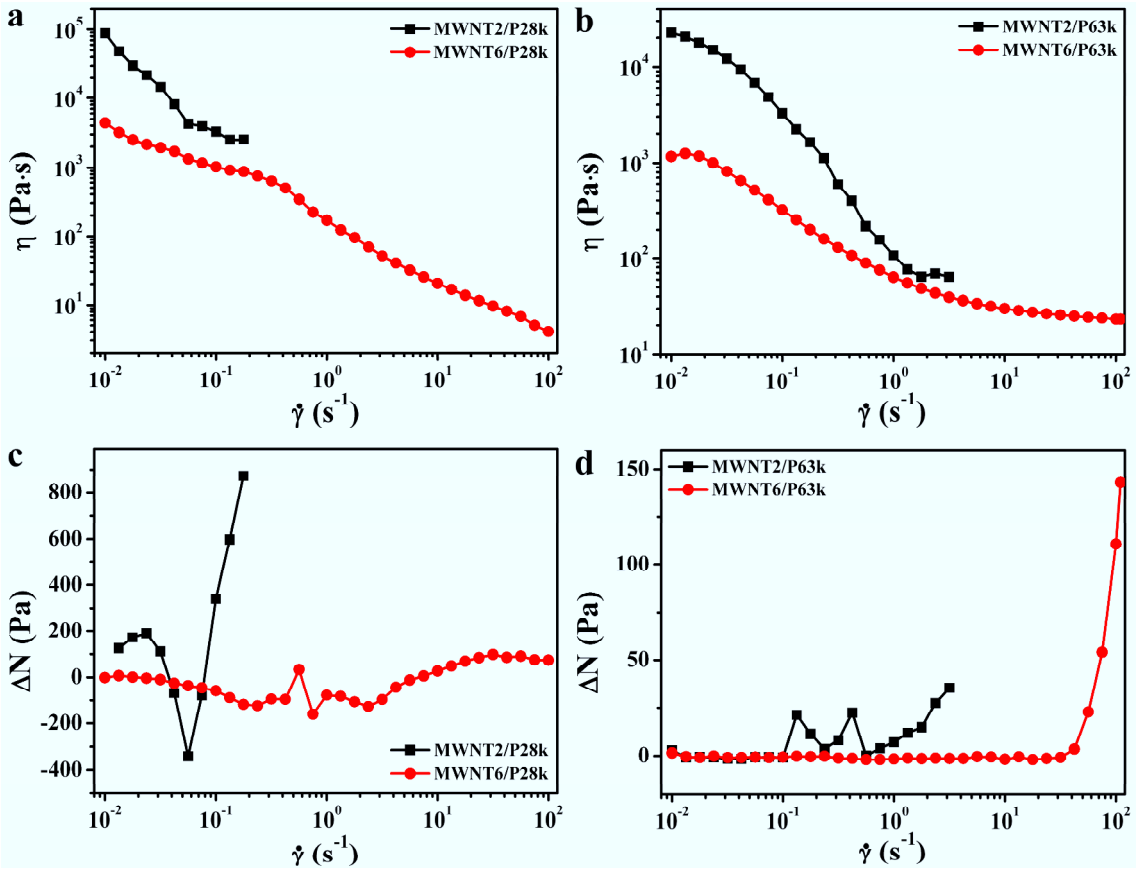

Fig. S9 Viscosity (η) and normal stress differences (ΔN) versus shear rate ($\dot{\gamma}$) for 16 wt % MWNT2/P28k and MWNT6/P28k (a and c); and 16 wt % MWNT2/P63k and MWNT6/P63k composites (b and d). The gap used in the rheological experiments is about 0.9 mm.

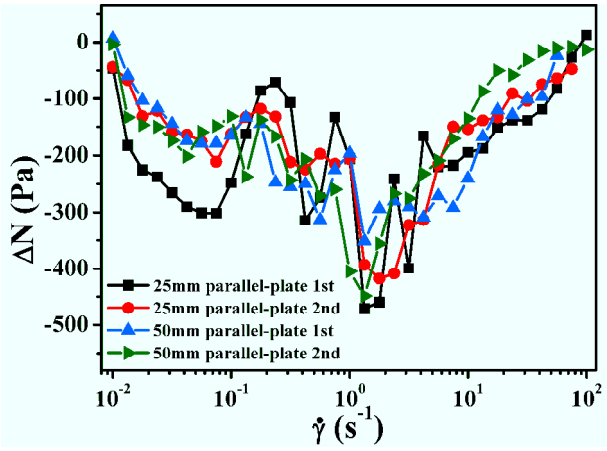

Fig. S10 Normal stress differences (ΔN) versus shear rate for 16 wt % MWNT2/P6k composites measured by different geometries.

Influence of concentration of MWNT on the rheological properties of MWNT/PDMS composites

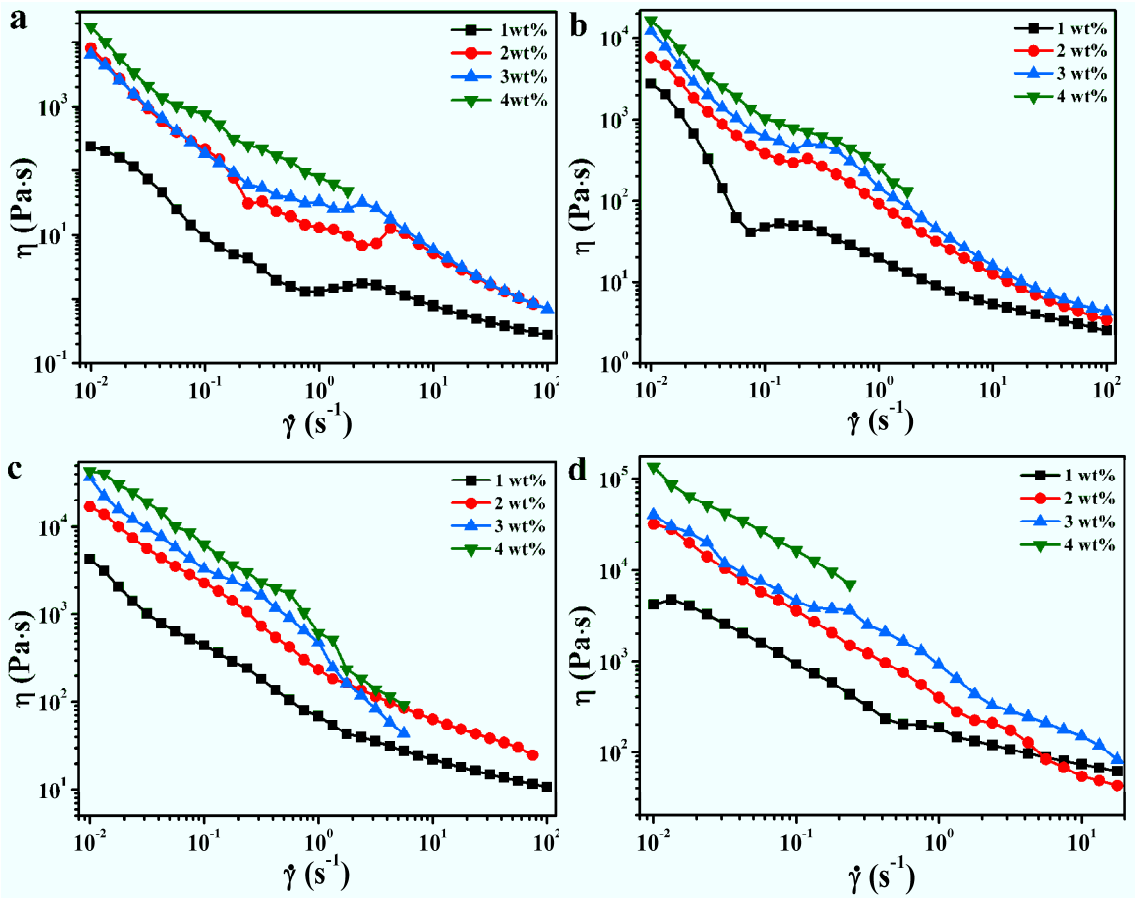

Fig. S11 Viscosity (η) versus shear rate ($\dot{\gamma}$) for MWNT0/P6k (a), MWNT0/P28k (b), MWNT0/P63k (c) and MWNT0/P117k composites (d) with different concentrations of MWNT0. The gap used in the rheological experiments is about 0.9 mm.

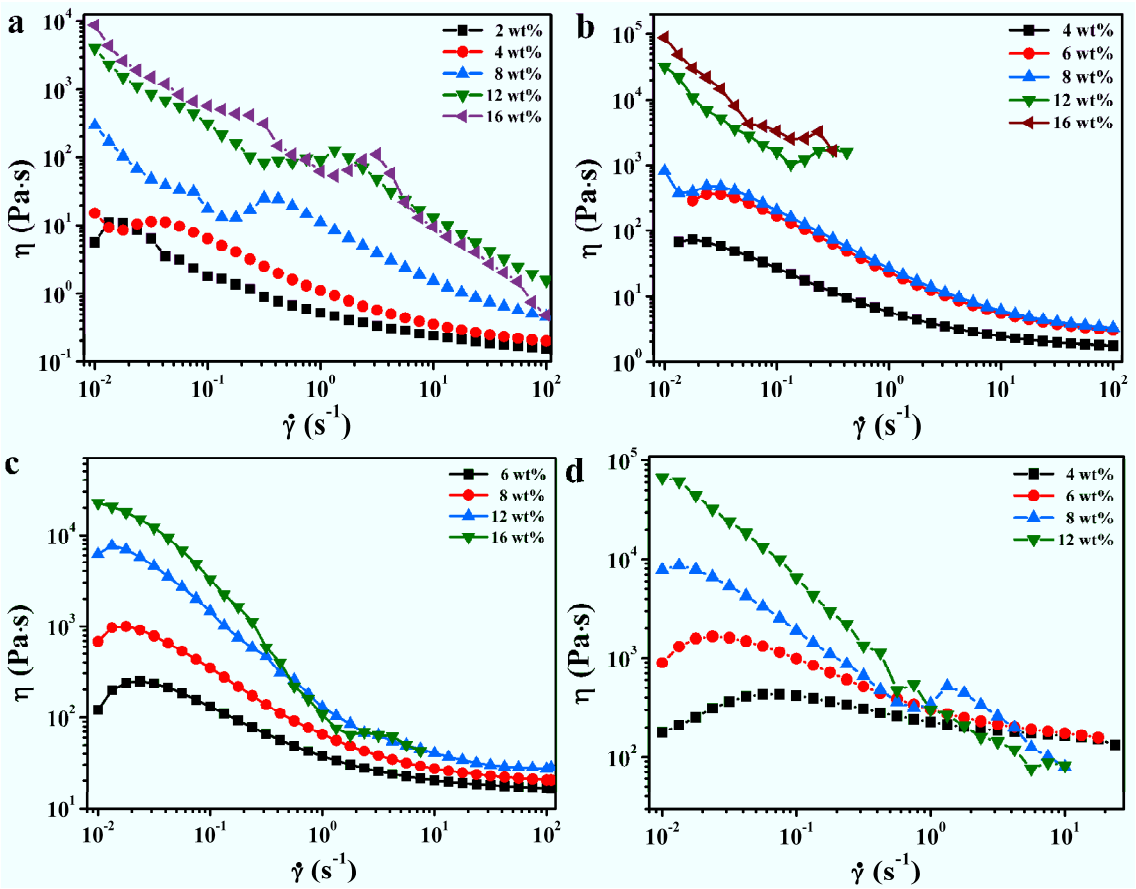

Fig. S12 Viscosity (η) versus shear rate ($\dot{\gamma}$) for MWNT2/P6k (a), MWNT2/P28k (b), MWNT2/P63k (c) and MWNT2/P117k composites (d) with different concentrations of MWNT2. The gap in the rheological experiments is about 0.9 mm.

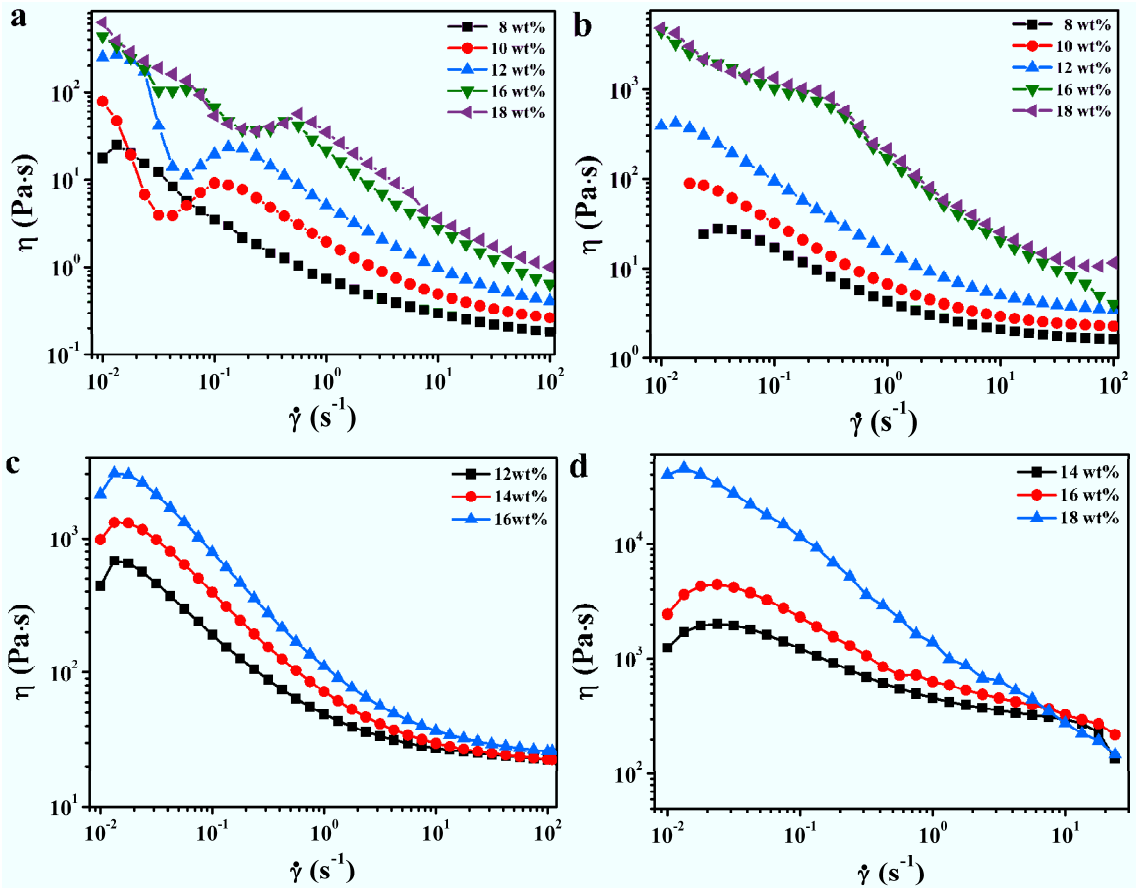

Fig. S13 Viscosity (η) versus shear rate ($\dot{\gamma}$) for MWNT6/P6k (a), MWNT6/P28k (b), MWNT6/P63k (c) and MWNT6/P117k composites (d) with different concentrations of MWNT6. The gap in the rheological experiments is about 0.9 mm.

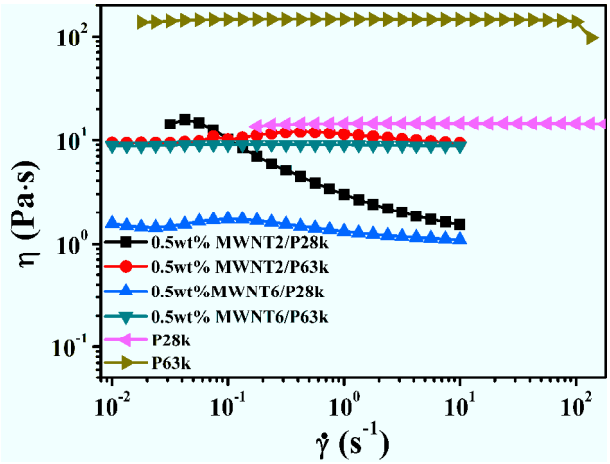

Fig. S14 Viscosity (η) versus shear rate ($\dot{\gamma}$) for MWNT2/P28k, MWNT2/P63k, MWNT6/P28k and MWNT6/P63k composites with 0.5 wt % MWNTs. The gap in the rheological experiments is about 150 μm.

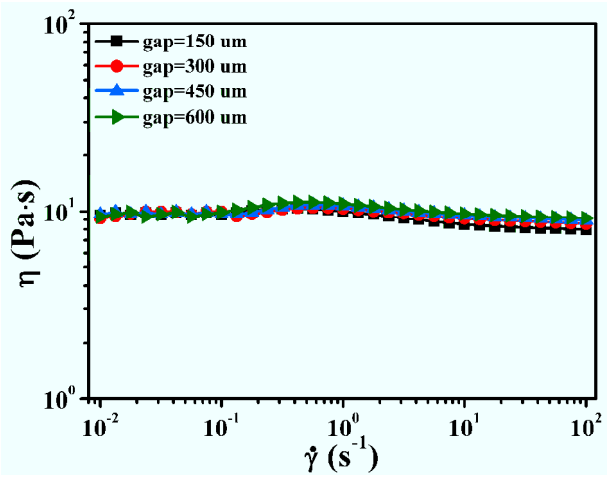

Fig. S15 Viscosity (η) versus shear rate ($\dot{\gamma}$) for 0.5 wt % MWNT2/P63k composites measured at different heights of gap.

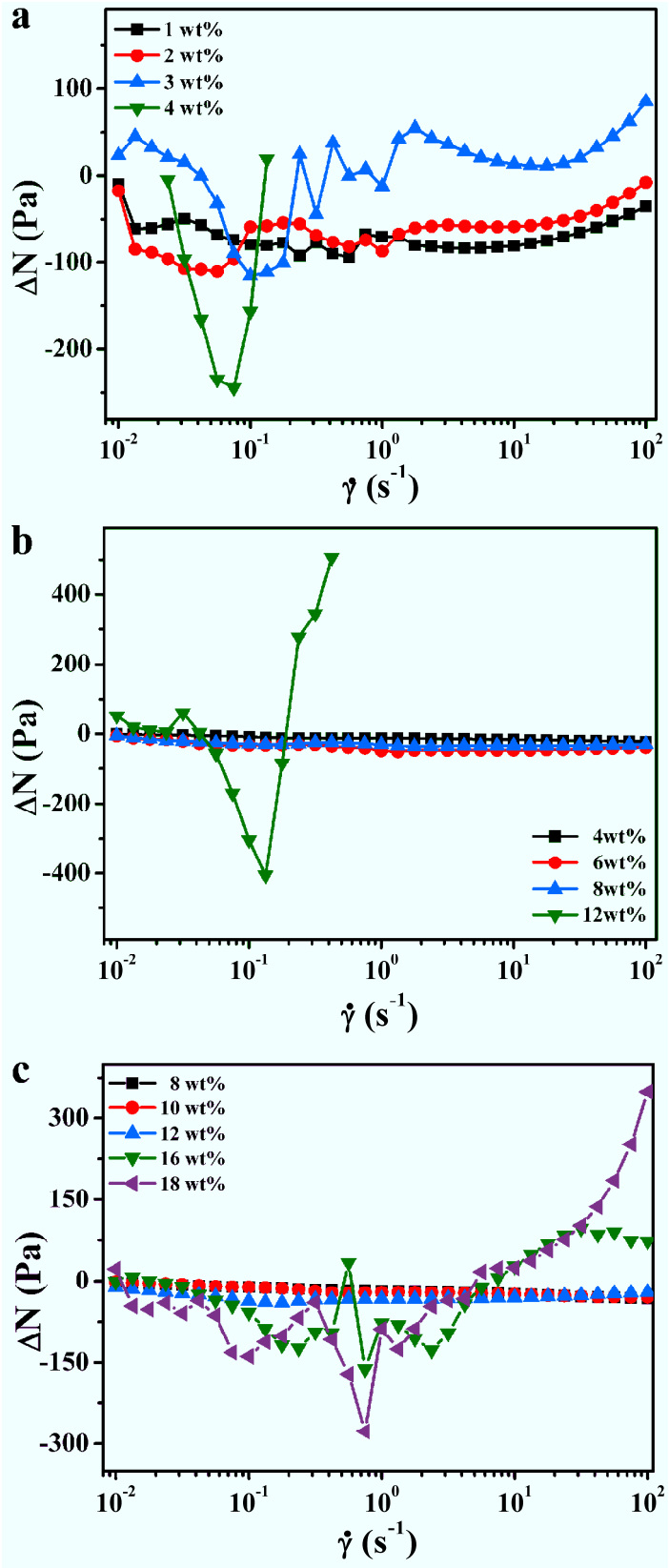

Fig. S16 Normal stress differences (ΔN) versus shear rate ($\dot{\gamma}$) for MWNT0/P28k (a), MWNT2/P28k (b) and MWNT6/P28k composites (c) with different concentrations of MWNT. The gap in the rheological experiments is about 0.9 mm.

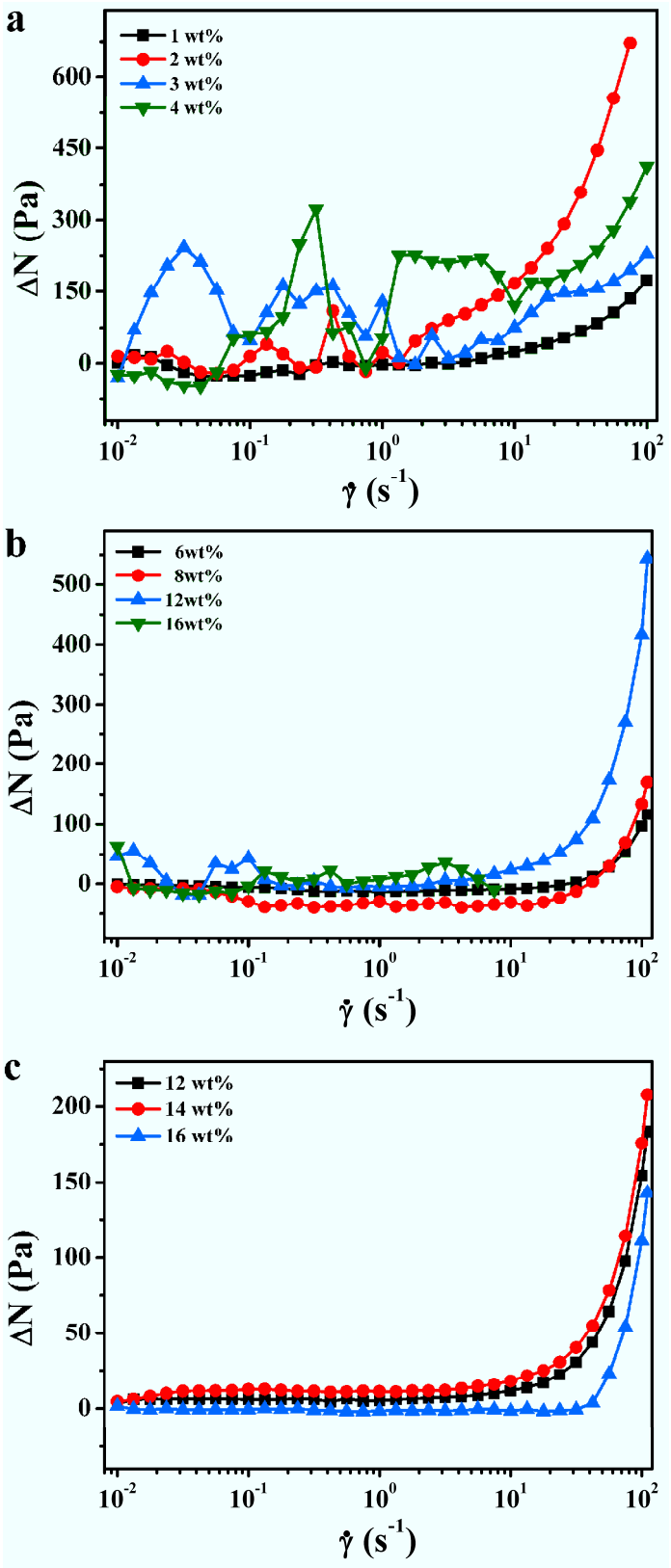

Fig. S17 Normal stress differences (ΔN) versus shear rate ($\dot{\gamma}$) for MWNT0/P63k (a), MWNT2/P63k (b) and MWNT6/P63k composites (c) with different concentrations of MWNT. The gap in the rheological experiments is about 0.9 mm.

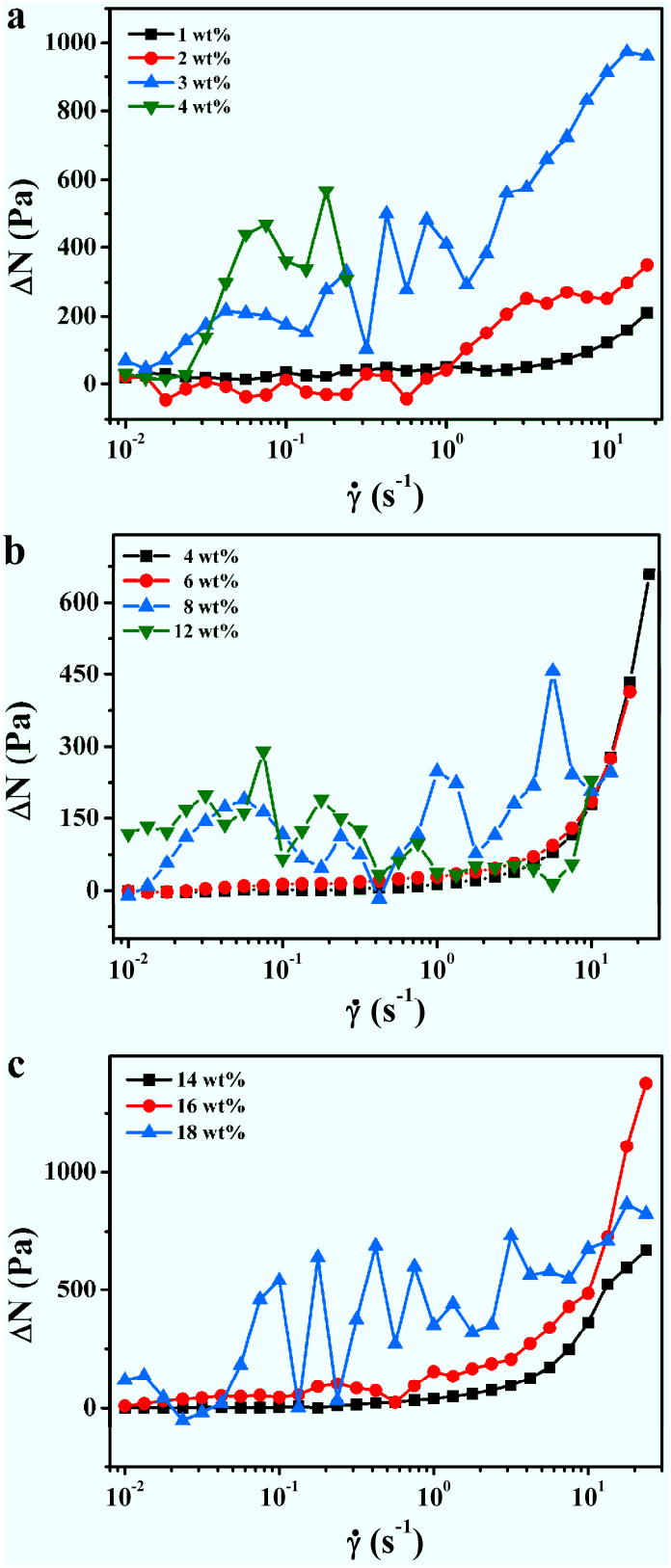

Fig. S18 Normal stress differences (ΔN) versus shear rate ($\dot{\gamma}$) for MWNT0/P117k (a), MWNT2/P117k (b) and MWNT6/P117k composites (c) with different concentrations of MWNT. The gap in the rheological experiments is about 0.9 mm.

Structural change under shear for MWNT/PDMS composites

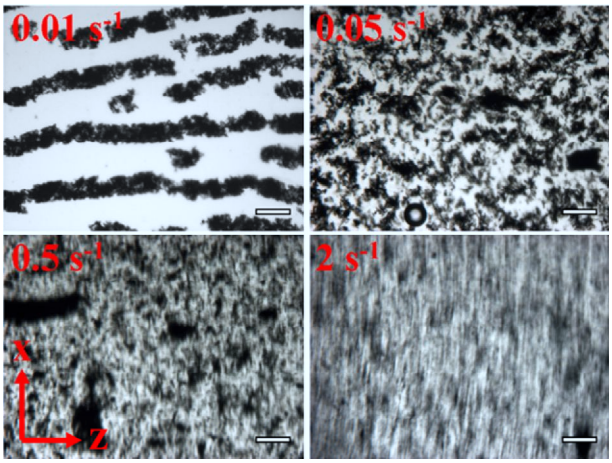

Fig. S19 Structures of 0.5 wt % MWNT0/P28k under different shear rates. The scale bars are 150 μm and the gap is 150 μm. Photos are taken after shearing for about 45 s.

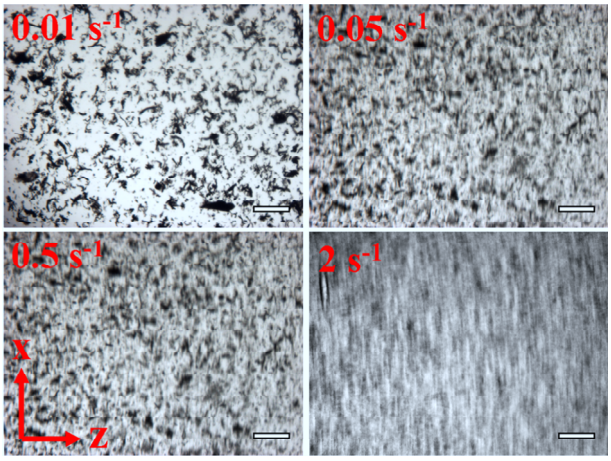

Fig. S20 Structures of 0.5 wt % MWNT0/P63k under different shear rates. The scale bars are 150 μm and the gap is 150 μm. Photos are taken after shearing for about 45 s.

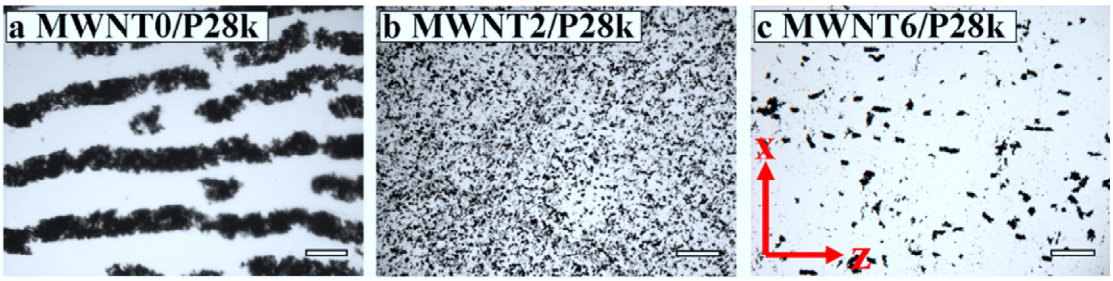

Fig. S21 Structures of 0.5 wt % MWNT0/P28k (a), 0.5 wt % MWNT2/P28k (b) and 0.5 wt % MWNT6/P28k (c) under a shear rate of 0.01 s^{-1}. The scale bars are 150 μm and the gap is 150 μm. Photos are taken after shearing for about 45 s.

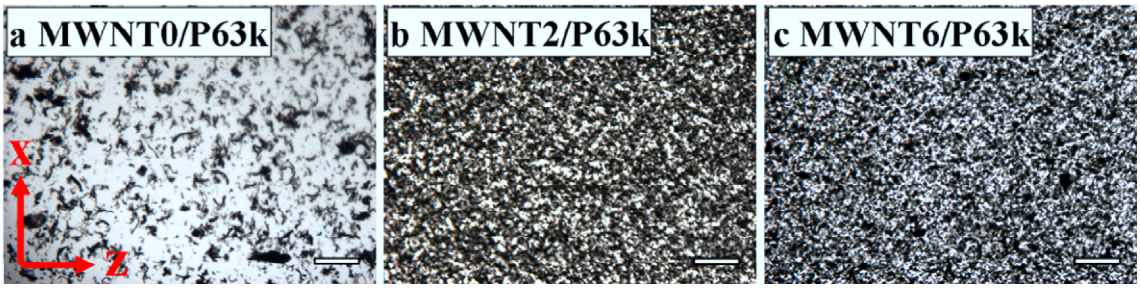

Fig. S22 Structures of 0.5 wt % MWNT0/P63k (a), 0.5 wt % MWNT2/P63k (b) and 0.5 wt % MWNT6/P63k (c) under a shear rate of 0.01 s^{-1}. The scale bars are 150 μm and the gap is 150 μm. Photos are taken after shearing for about 45 s.

Fig. S23 Structures of 0.5 wt % MWNT0/P117k (a), 0.5 wt % MWNT2/P117k (b) and 0.5 wt % MWNT6/P117k (c) under a shear rate of 0.01 s^{-1}. The scale bars are 150 μm and the gap is 150 μm. Photos are taken after shearing for about 45 s.

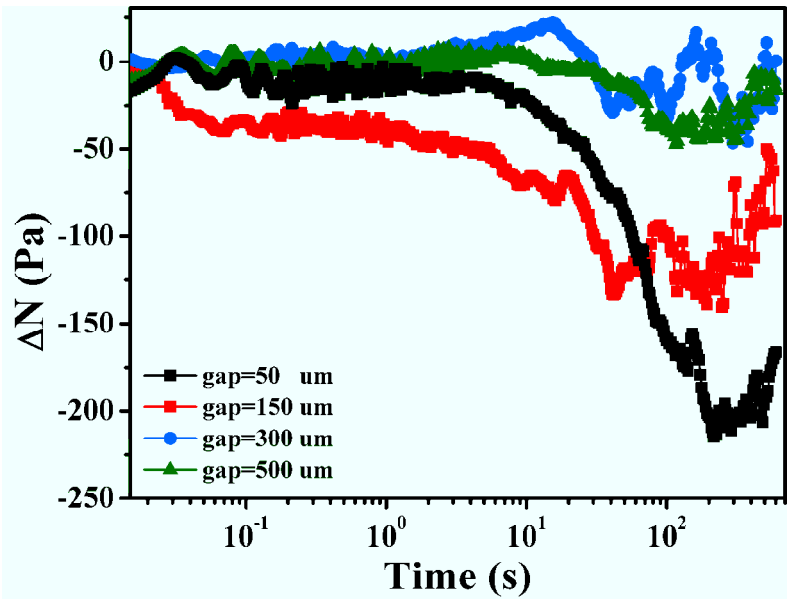

Fig. S24 Normal stress differences (ΔN) versus time at $\dot{\gamma}$ = 0.01 s^{-1} for 1 wt % MWNT0/P28k under the gap of 50, 150, 300 and 500 μm.

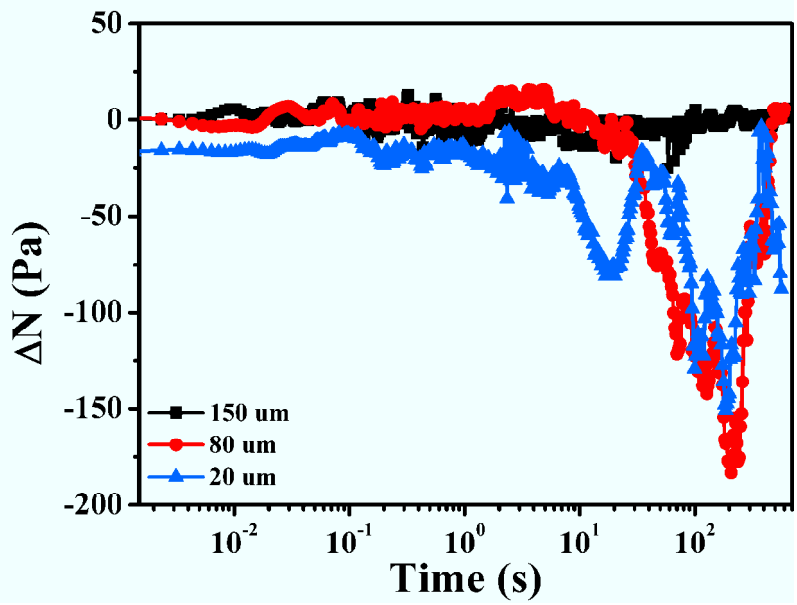

Fig. S25 Normal stress differences (ΔN) versus time at $\dot{\gamma}$ = 0.01 s^{-1} for 0.5 wt % MWNT0/P63k under the gap of 150, 80 and 20 μm.

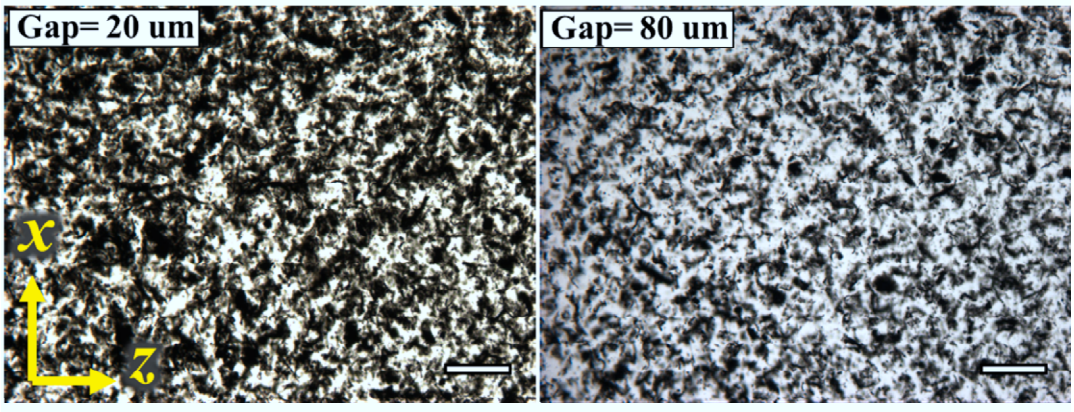

Fig. S26 Structures of 0.5 wt % MWNT0/P117k with the gap of 80 and 20 μm at a shear rate of 0.01 s^{-1}. The scale bars are 150 μm. Photos are taken after shearing for 150 s.

Average aggregate size of MWNTs in MWNT/PDMS composites

Table S1 Mean aggregate size of 0.5 wt % MWNT/PDMS composites

MWNT0/PDMS	R_0 (μm)	MWNT2/PDMS	R_0 (μm)	MWNT6/PDMS	R_0 (μm)
MWNT0/P6k	96.2	MWNT2/P6k	26.5	MWNT6/P6k	19.0
MWNT0/P28k	66.4	MWNT2/P28k	22.4	MWNT6/P28k	16.8
MWNT0/P63k	48.9	MWNT2/P63k	19.9	MWNT6/P63k	9.1
MWNT0/P117k	30.4	MWNT2/P117k	9.7	MWNT6/P117k	7.5

Table S2 Mean aggregate size of 2 wt % MWNT2/PDMS and 4 wt % MWNT6/PDMS composites

2 wt % MWNT2/PDMS	R_0 (μm)	4 wt % MWNT6/PDMS	R_0 (μm)
2 wt % MWNT2/P6k	68.4	4 wt % MWNT6/P6k	50.3
2 wt % MWNT2/P28k	58.1	4 wt % MWNT6/P28k	44.6
2 wt % MWNT2/P63k	27.0	4 wt % MWNT6/P63k	36.3
2 wt % MWNT2/P117k	20.1	4 wt % MWNT6/P117k	17.6

Steady shear results of pure PDMS

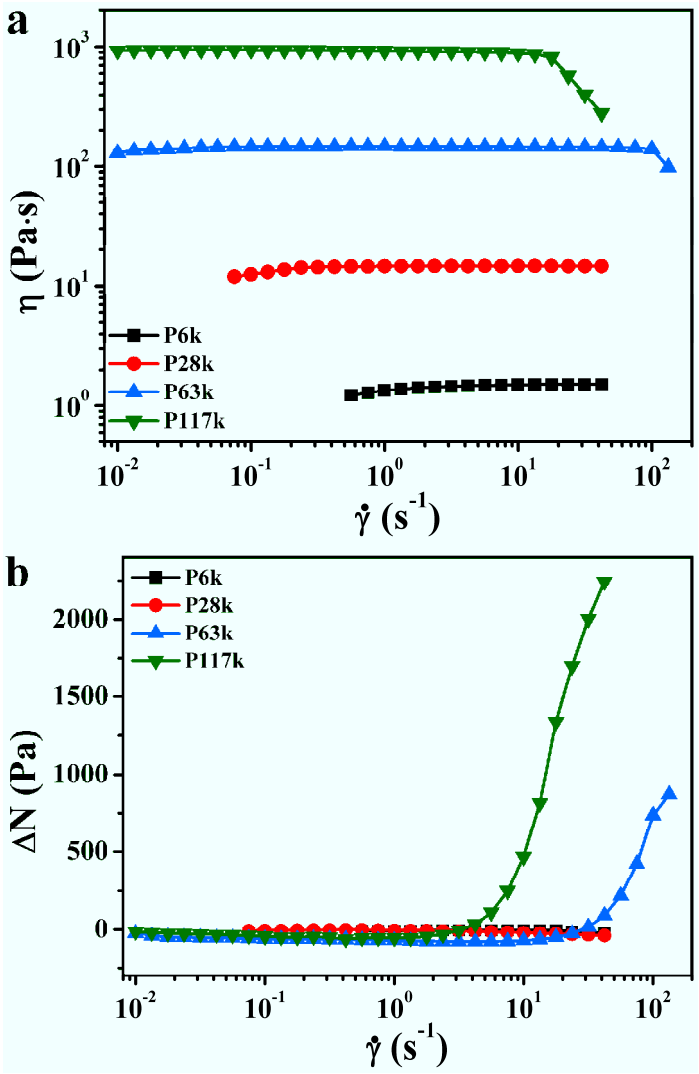

Fig. S27 Viscosity (a) and normal stress differences (b) versus shear rate for pure PDMS with different molecular weights. The gap in the rheological experiments is about 0.9 mm.

S1 K. Nishinari, *Prog. Colloid Polym. Sci.*, 2009, **136**, 87–94.

S2 R. Krishnamoorti and E. P. Giannelis, *Macromolecules*, 1997, **30**, 4097–4102.

S3 A. K. Kota, B. H. Cipriano, M. K. Duesterberg, A. L. Gershon, D. Powell, S. R. Raghavan and H. A. Bruck, *Macromolecules*, 2007, **40**, 7400–7406.