Supplementary information

Synthesis of NIPA core/NIPMA shell microgels

NIPA core/NIPMA shell microgels (denoted by **(N)-NM**, Table 2) were synthesized as follows: First, NIPA (3.34 g, 150 mM) was dissolved in 200 mL of water. The monomer solution was then poured into a three-neck, round-bottom flask equipped with a mechanical stirrer, a condenser, and nitrogen gas inlet. The solution was bubbled for 30 min with nitrogen gas to purge oxygen at 40 °C. Under a stream of nitrogen and with constant stirring at 250 rpm, 29.7 μL of *N*,*N*,*N*',*N*'-tetramethylethylenediamine (TEMED) and the initiator KPS dissolved in 10 mL of water was injected into the flask to start the polymerization. After 3 h, temperature was increased to 70 °C. After another 1 h, a mixture of NIPMA, BIS, and 200 mL of water was poured into the flask to form NIPMA-based shell on NIPA-based core. Here, The total monomer concentration and the comonomer ratio, (1-*X*:*X*) (NIPMA:BIS) were varied as shown in Table 2. After another 4 h, the microgel suspension was cooled to room temperature. The microgels were purified by centrifugation/redispersion with water twice using a RCF of 50000g, and by means of dialysis. After that, the suspensions were concentrated by centrifugation (50000g) to obtain the microgel pastes.

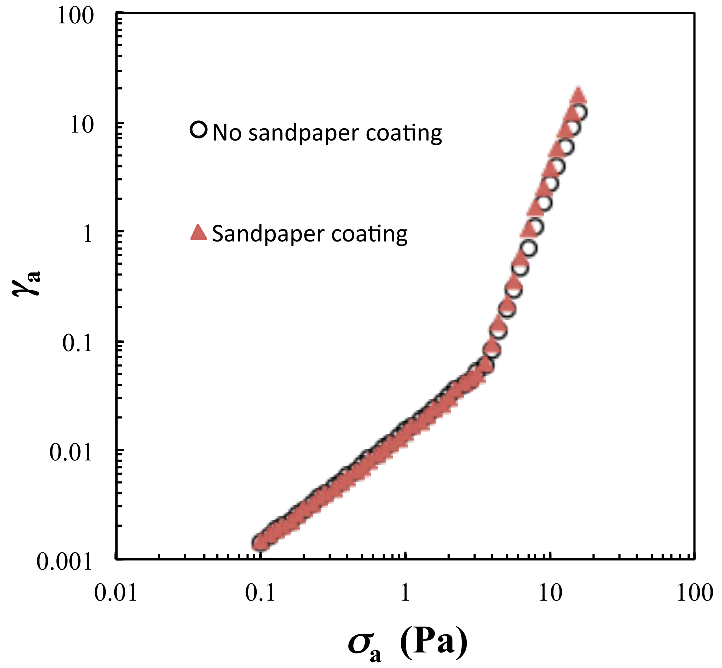

Fig. S1: Comparison of the σ_a-γ_a data obtained using the metal plate whose surface is coated with or without waterproof sandpaper for a dense suspension of N-1-710 with $c = 4.31 \times 10^{-2}$ g/ml. No appreciable difference is observed.

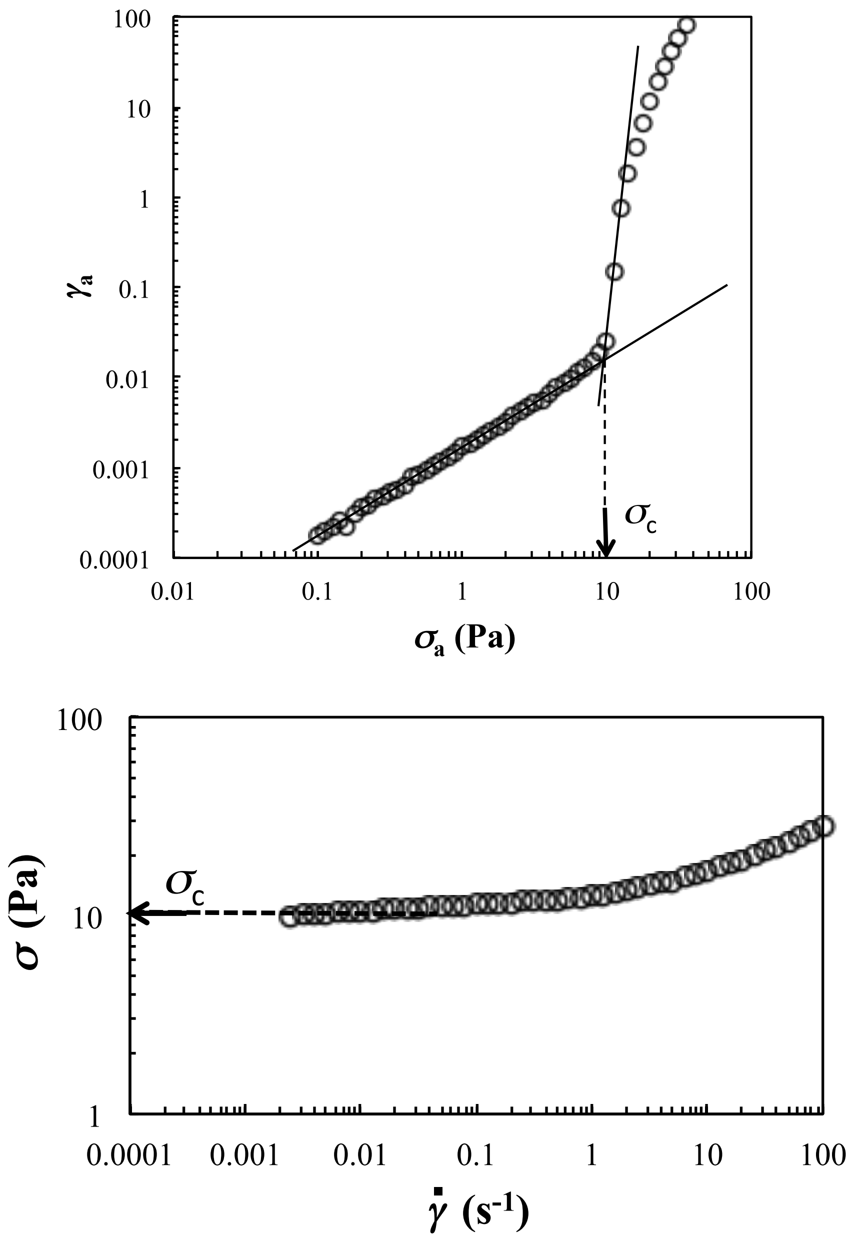

Fig. S2: Comparison of the values of yield stress (σ_c) evaluated from (upper) the oscillatory experiments using stress amplitude as a variable at an angular frequency of 1 s^{-1}, and (lower) the steady-state flow experiments for a dense microgel suspension. The same value of σ_c (10 Pa) is obtained from these separate experiments.

Table S1. Concentration (c), apparent volume fraction (ϕ_{eff}), equilibrium shear modulus (G_0), yield stress (σ_c) and yield strain (γ_c) for the suspensions of the binary mixtures of NM-2-1600 and NM-5-750, the core-shell microgels, the copolymer microgels.

Microgel	c(g/ml)	ϕ_{eff}	G_0(Pa)	σ_c(Pa)	$\gamma_c \times 10^{-2}$
NM-2-1600/NM-5-750 1/1 blend	6.30×10^{-2}	-	1.0×10^{2}	2.7×10^{0}	2.6
NM-2-1600/NM-5-750 3/1 blend	6.30×10^{-2}	-	7.7×10^{1}	2.1×10^{0}	2.8
NM-2-1600/NM-5-750 1/3 blend	6.30×10^{-2}	-	1.3×10^{2}	3.1×10^{0}	2.5
(N)-NM-5-100-1100	5.11×10^{-2}	0.890	1.0×10^{2}	2.7×10^{0}	2.7
(N)-NM-5-70-1200	3.09×10^{-2}	0.928	1.2×10^{2}	3.2×10^{0}	2.6
(N)-NM-10-100-1100	1.15×10^{-1}	0.893	8.9×10^{2}	2.2×10^{1}	2.5
N49.5-NM49.5-1-920	5.14×10^{-2}	1.29	3.9×10^{2}	1.3×10^{1}	3.4
	4.65×10^{-2}	1.17	1.1×10^{2}	4.3×10^{0}	3.8
	4.02×10^{-2}	1.01	3.0×10^{1}	1.1×10^{0}	3.6
	3.47×10^{-2}	0.870	1.1×10^{1}	4.4×10^{-1}	4.0

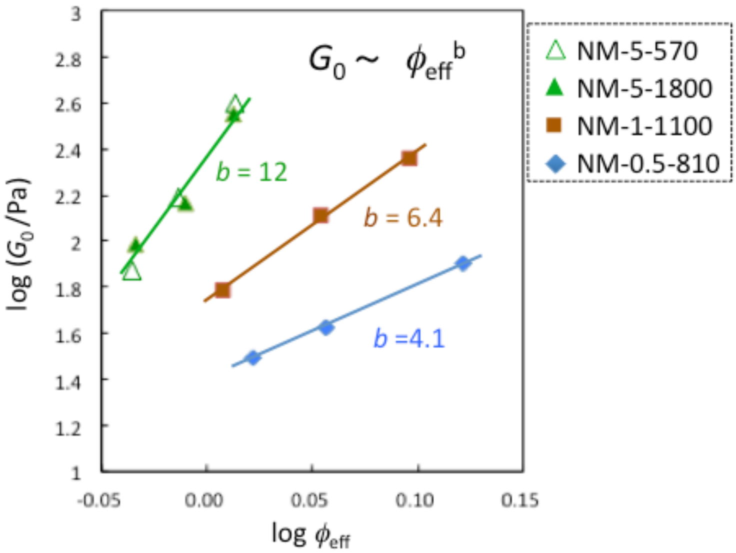

Fig. S3: Double logarithmic plots of equilibrium shear modulus (G_0) and apparent volume fraction of particle (ϕ_{eff}) for the dense pastes of PNIPMA microgels.