



Concepts and conflicts in nanoparticles reinforcement to polymers beyond hydrodynamics



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ABSTRACT

With substantial progresses in reinforcement mechanism of nanoparticle filled polymers (NPPFs) beyond the hydrodynamic limit, now is the time to reconsider this topic characterized by emergence of a universal solid-like rheology at nanoparticle contents above a critical value in relation to the heterogeneously retarded polymer dynamics and structured

Abbreviations: AFM, atomic force microscopy; α , an exponent representing for dynamics of the “filler phase”; $a(\varphi)$, horizontal shift factor for constructing $G^*(\omega, \varphi)$ and $G(t, \varphi)$ master curves; $A_f(\varphi)$, strain simplification factor; β , scaling exponent; $1/b(\varphi)$, vertical shift factor or constructing $G^*(\omega, \varphi)$ and $G(t, \varphi)$ master curves; CB, carbon black; CCA, cluster-cluster aggregation; $C(r)$, radially averaged autocorrelation function; δ , a measure of the number of deformable links in a strand; d_f , fractal dimension; d_{fb} , fractal dimension of cluster backbones; DLCA, diffusion-limited cluster aggregations; d_{\min} , minimum spacing between nearest neighbor nanoparticles; $d_{0\text{ inter}}$, interacting distance between primary cluster; ε , a measure of the bendability of the link; EPDM, ethylene-propylene-diene monomer rubber; EVA, ethylene-vinyl acetate; Δ , length parameter; δ , phase lag between strain and stress; η , viscosity; $[\eta]$, intrinsic viscosity; η_r , relative viscosity; η_s , viscosity of suspending fluid; $\eta^*(\omega, \varphi)$, complex viscosity; $\eta_0(\varphi)$, zero-shear viscosity of NPPFs; φ , particle volume fraction; φ_c , critical threshold for fractal network formation; φ^{**} , glass transition limit; φ_e , effective filler volume fraction; φ_{ec} , critical effective concentration; φ_{eb} , volume fraction of aggregates within the branches; φ_{fract} , volume fraction of fractals; φ_m , maximum packing limit of nanoparticles; φ^* , nanoparticle clustering threshold; γ , strain amplitude; $G_c(\varphi)$, $G'-G''$ crossover; G_{dis} , modulus of dispersed filler; $G'_{f0}(\varphi)$, constant representing viscous contribution of the filler phase; $G'_{\text{gel},0}$, viscous component of the gel stiffness; $G''_m(\omega)$, viscous modulus of the suspending medium; $G_c(\varphi)$, equilibrium modulus at time infinity; g_i , modulus of Maxwellian units; $g(\varphi)$, ω -independent modulus amplitude function; $G'_{f1}(\varphi)$, storage modulus at $\omega = 1 \text{ rad s}^{-1}$; $G'_{\text{gel},0}$, elastic component of the gel stiffness; $G^*(\omega, \varphi)$, storage modulus at frequency ω for NPPFs with particle volume fraction φ ; G_{net} , modulus of percolated filler network; GNPs, graphite nanoplatelets; G_N^0 , plateau modulus in the regime of matrix entanglement; $G^*(\omega, \varphi)$, complex modulus; $G_f^*(\omega, \varphi)$, complex modulus of the filler network bridged by polymer chains; $G_m^*(\omega)$, complex modulus of the matrix; $G(t, \varphi)$, linear relaxation modulus; $G_0(\varphi)$, modulus plateau for NPPFs with filler volume fraction φ ; H_{dis} , relative modulus of dispersed filler and matrix with/without isolated aggregates; $H(\varphi)$, φ -dependent hydrodynamic amplification factor; H_s , thickness of the slice used for TEM observation; $H(\tau)$, relaxation time spectrum; H_{net} , relative modulus of percolated filler network and matrix; k , shape factor of loosely packed structure of the fractal clusters; κ , compactness or internal aggregate volume fraction; LLDPE, linear low density polyethylene; m , relaxation exponent lying between 0 and 1; m_e , relaxation parameters; m_g , relaxation parameters; MWCNTs, multi-walled carbon nanotube; n , exponent defining the liquid- ($n = 1$) to solid-like ($n = 0$) transition; N_{agg} , number of particles per aggregate; NPPFs, nanoparticle filled polymers; NR, natural rubber; ω , frequency; $\omega_c(\varphi)$, $G'-G''$ crossover frequency; P2VP, poly(2-vinylpyridine); PB, polybutadiene; PBO, poly(butylene oxide); PDMS, polydimethylsiloxane; PEMA, poly(ethyl methacrylate); PE-g-MA, maleic anhydride grafted polyethylene; PEO, poly(ethylene oxide); PEP, poly(ethylene-*alt*-propylene); phr, parts per hundred rubber; PI, polyisoprene; $\Phi(\varphi)$, theoretical volume fraction of percolated filler network; PMMA, poly(methyl methacrylate); PP, polypropylene; PS, polystyrene; PTHF, polytetrahydrofuran; PVAc, poly(vinyl acetate); q , wave vectors; R , diameter of primary particle; R_a , diameter of aggregates; $R'_{f(\varphi)}$, scaling factor representing viscous contributions of the “filler phase”; $I(q)/\varphi$, reduced scattering intensities; R_g , radius of gyration of polymer; R_{g0} , radius of gyration of unperturbed polymer; RLCA, reaction-limited cluster aggregations; $R'_{f(\varphi)}$, scaling factor representing elastic contribution of the “filler phase”; ρ_{net} , filler network density; SANS, small-angle neutron scattering; SAXS, small-angle X-ray scattering; SBR, styrene-butadiene rubber; σ_f , stress due to interactions of attractive nanoparticles dispersed in the matrix; $\Theta(\varphi_e)$, scaling function; σ_m , viscoelastic stress arising from micro-flow of the matrix around flocs; $S(\omega)$, φ -independent shape function; S_{sa} , specific surface area; t , time; $\tan \delta$, mechanical loss; τ , relaxation time; τ_i , relaxation time of Maxwellian units; τ_{max} , disentanglement time of a full chain; τ_0 , the shortest relaxation time of entanglement segments, or short relaxation time which characterizes the structural building block of the gel; TCS, time-concentration superposition; T_{li} , thickness of the glassy layer; TMDSC, temperature-modulated differential scanning calorimetry; T_N , mean-field “noise temperature” related to the phase lag; TEM, transmission electron microscopy; TEMT, transmission electron microtomography; T_g , glass transition temperature; TTS, time-temperature superposition (TTS); x , exponent; ζ , a measure of strength links between the flocs; y , exponent; z , exponent.

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filler network. A comprehensive survey is presented herein to report the cluster-cluster aggregation model, and jamming, percolation and soft colloidal dynamics theories and their applications in NPPFs in relation to nanoparticle reinforcement of polymers beyond hydrodynamics. Methods for separating the filler contribution to the reinforcement within the frame work of two-phase models are highlighted and three main kinds of theoretical models (cluster-cluster aggregation model, jamming theory, and rigidity percolation theory) are discussed for addressing the conflicts of concepts in experiments. Many years research practice shows that there is still lack of a credible relation describing the reinforcement with respect to the network structure (nanoparticle dispersity) mediated by microscopic interaction and sample preparation. Furthermore, viscoelasticity of the filler phase is discussed according to two-phase models and beyond. Finally several open challenges are remarked.

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