Multi-Scale Effects on Deformation Mechanisms of Polymer Nanocomposites: Experimental Characterisation and Numerical Study

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Abstract

In order to make much stiffer, light weight and high performance material products, polymer nanocomposites play an emerging role in the material innovation. Unlike other thermoplastics, polymer nanocomposites are fabricated by introducing a small amount of solid nano-scale fillers (normally less than 5 wt%) such as nanoclay, carbon nanotubes or nanofibres into a plastic resin to dramatically enhance its stiffness, strength and thermal properties. The difference between nanocomposites and conventional fibre composites is that the added fillers are extremely small, only one-millionth of a millimetre thick, and provide a much larger interface area per unit volume for greatly improving the interfacial bonding effect between nanofillers and the polymer matrix.

More importantly, polypropylene (PP)/clay nanocomposites have quite a high potential to form such innovative materials and replace the conventional plastics in many automotive and packaging applications. Nevertheless, the growth of PP/clay nanocomposites faces an obstacle of hydrophobic polymer's low interactions with hydrophilic clay. Maleic anhydride (MA) grafted PP (MAPP), commonly used as a compatibiliser, has been proven to facilitate a good clay dispersion within the PP matrix through its functionalised MA groups. But despite the great attention from the manufacturers and researchers in recent years, commercial PP/clay nanocomposites with reliable material properties are still limited in availability. The major problem stems from the complex influences of the material selection and processing methods.

The present work developed a comprehensive approach from the material formulation and processing, experimental characterisation to the numerical modelling of PP/clay nanocomposites based on the finite element analysis (FEA) of micro/nanostructures. Initially, effects of the material selection including the clay type and content, MAPP content and PP matrix viscosity were investigated for the mechanical property enhancement of PP/clay nanocomposites. These nanocomposites were prepared using twin screw extrusion and injection moulding processes with a well-known Taguchi design of experiments (DoE) method in order to statistically detect the significant factors for influencing their mechanical properties. The preferred material formulations were then determined by Pareto analysis of variance (ANOVA) with the technical and economic considerations. The fundamental material characterisation was also conducted on those formulated nanocomposites using X-ray diffraction (XRD), scanning electron microscopy (SEM), transmission electron microscopy (TEM), differential scanning calorimetry (DSC) and dynamic mechanical thermal analysis (DMTA). Overall mechanical properties of neat PP and corresponding nanocomposites were determined by the general tensile, flexural and impact tests. Finally, computational models were established by implementing both the representative volume element (RVE) technique and innovative object-oriented finite element (OOF) analysis to predict the tensile moduli of PP/clay nanocomposites in comparison to the experimental data and available composites theoretical models.

Dedication

To

My parents Fengzhang Dong & Jinglan Wang

and

My wife Ruixue (Marie) Ma

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Nomenclatures

A	Strain concentration tensor of polymer composites
A_0	Cross section area of 3×3 RVE domain
A_i	Components of Eshelby tensor (i=0, 1,….., 5)
A_T	Area of mesh triangle T in shape energy function
a	Length of single RVE domain
$a_i(T)$	Area fraction for each mesh triangle T including pixels of a given gray scale i in homogeneity energy function
B	Stress concentration tensor of polymer composites
b	Width of single RVE domain
b_0	Width of injection moulded tensile sample
C	Stiffness tensor of polymer composites
C^f	Elastic stiffness tensor of fillers in polymer composites
C^m	Elastic stiffness tensor of matrix in polymer composites
c	Lateral distance between two clay platelets in RVE models
D	Screw diameter
d	Interlayer spacing between two layer planes in X-ray diffraction
d_{MMT}	Silicate layer thickness (i.e. interlayer spacing of pure MMT)
d_i	Galley thickness of clay silicates
d_0	Diameter/width of dispersed fillers in polymer composites
d_{001}	Interlayer spacing from (001) basal reflection peak
$\boldsymbol{E}$	Total energy function parameter

E_{11}	Longitudinal elastic modulus of polymer composites
E_{22}	Transverse elastic modulus of polymer composites
E_{33}	Elastic modulus of polymer composites along axis 3 loading direction
E_{MMT}	Elastic modulus of pure MMT
E_c	Young's modulus of polymer composites
E_{eff}	Effective tensile modulus in OOF models
E_f	Young's modulus of fillers in polymer composites
$E_{gallery}$	Elastic modulus of the interlayer (intragallery) material in clay silicates
$\boldsymbol{E_{hom}}$	Homogeneity energy function parameter
E_m	Young's modulus of matrix in polymer composites
E_p	Elastic modulus of stacks of layered intercalated clay platelets
E_{ran-3D}^{fibres}	Elastic modulus of fibre reinforced composites in the 3-D orientation
$E_{ran-3D}^{platelets}$	Elastic modulus of platelet reinforced composites in the 3-D orientation
$\boldsymbol{E_{shape}}$	Shape energy function parameter
E'	Elastic (storage) modulus in DMTA
E''	Loss modulus in DMTA
$E_{//}$	Composite modulus parallel to the major axis of fillers
$E_{\perp}$	Composite modulus perpendicular to the major axis of fillers
F_i	Nodal force at node i
f_s	Staggering factor in RVE models
g	$g = \frac{\pi}{2}\alpha$
ΔH_f^0	Heat of fusion of pure crystalline PP

ΔH_m	Heat of fusion of the PP matrix
k	Modulus ratio of interface in Takayanagi model
L	Length of dispersed fillers in polymer composites
L/D	Screw length to diameter ratio
L/t (L/d_0)	Aspect ratio of dispersed fillers in polymer composites
L_0	Length of injection moulded tensile sample
L_T	Perimeter of mesh triangle T in shape energy function
L_x	Horizontal dimension of OOF models
L_y	Vertical dimension of OOF models
ΔL	In-plane displacement in OOF models
ΔL_0	Offset between clay platelets for the stack rows
Λ	$\Lambda = (1-\phi_f)\left[\frac{3(\alpha^2+0.25)g-2\alpha^2}{\alpha^2-1}\right]$
N	Number of categories of the pixel selection in homogeneity energy function
n	Integral number in Bragg's law
n_0	Number of samples in each trial of Taguchi DoE
n'	Set number of boundary nodes under the nonzero displacement condition in OOF modelling
S	Average compliance of polymer composites
S/N	Signal to noise ratio in Taguchi DoE
S^f	Compliance tensor of fillers in polymer composites
S^m	Compliance tensor of matrix in polymer composites
T_c	Crystallisation temperature
T_g	Glass transition temperature
T_m	Melting temperature
t	Thickness of dispersed fillers in polymer composites
t_0	Thickness of injection moulded tensile sample
$tan\delta$	Loss tangent in DMTA
U_x	Elastic strain applied on the boundaries of OOF models

	in the x direction
U_y	Elastic strain applied on the boundaries of OOF models in the y direction
V	Representative averaging volume of polymer composites
V_f	Volume of fillers in polymer composites
V_m	Volume of matrix in polymer composites
W_f	Clay weight fraction
X_c	Degree of crystallinity
y	Measured response value in Taguchi DoE
α	Inverse aspect ratio of dispersed fillers in polymer composites
$\boldsymbol{\alpha}, \boldsymbol{\beta}$	Dimensions in the transverse and longitudinal directions for interface in Takayanagi model
$\boldsymbol{\alpha_0}$	Tunable parameter in total energy function
ε_0	Applied strain
ε^f	Infinitesimal strain tensor of fillers in polymer composites
ε^m	Infinitesimal strain tensor of matrix in polymer composites
$\bar{\varepsilon}$	Volume-average strain of polymer composites
$\bar{\varepsilon}_f$	Volume-average strain of fillers in polymer composites
$\bar{\varepsilon}_m$	Volume-average strain of matrix in polymer composites
$\varepsilon(x)$	Strain field in polymer composites
ε_{xx}	Elastic tensile strain in the contour plots of OOF models
ζ	Shape parameter depending on filler geometry and loading direction
η	$\eta = \dfrac{\dfrac{E_f}{E_m} - 1}{\dfrac{E_f}{E_m} + \zeta}$
θ	Measured diffraction angle
λ	Wave length of X-ray radiation

λ, φ	Dimensions in the transverse and longitudinal directions for dispersed phase in Takayanagi model
ξ	$\xi = \phi_f + \frac{E_m}{E_f - E_m} + 3(1-\phi_f)\left[\frac{(1-g)\alpha^2 - \frac{g}{2}}{\alpha^2 - 1}\right]$
ρ_f	Density of clay particles
ρ_m	Density of the PP matrix
σ	Resulting axial stress
σ^f	Total stress tensor of fillers in polymer composites
σ^m	Total stress tensor of matrix in polymer composites
$\overline{\sigma}$	Volume-average stress of polymer composites
$\overline{\sigma}_f$	Volume-average stress of fillers in polymer composites
$\overline{\sigma}_m$	Volume-average stress of matrix in polymer composites
$\sigma(x)$	Stress field in polymer composites
σ_{xx}	Uniaxial stress in the contour plots of OOF models
τ	Thickness of interfacial region in Takayanagi model
ν_{MMT}	Poisson's ratio of pure MMT
$\nu_{gallery}$	Poisson's ratio of the interlayer (intragallery) material of clay silicates
ν_m	Poisson's ratio of matrix in polymer composites
ν_p	Poisson's ratio of stacks of layered intercalated clay platelets
ϕ_{MMT}	Volume fraction of silicate layers in the stack
ϕ_f	Volume fraction of fillers in polymer composites
ϕ_{galley}	Volume fraction of gallery space (i.e. interlayer area)
ϕ_m	Volume fraction of matrix in polymer composites