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Effective viscoelastic behavior of particulate polymer composites at finite concentration *

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Abstract Polymeric materials usually present some viscoelastic behavior. To improve the mechanical behavior of these materials, ceramics materials are often filled into the polymeric materials in form of fiber or particle. A micromechanical model was proposed to estimate the overall viscoelastic behavior for particulate polymer composites, especially for high volume concentration of filled particles. The method is based on Laplace transform technique and an elastic model including two-particle interaction. The effective creep compliance and the stress and strain relation at a constant loading rate are analyzed. The results show that the proposed method predicts a significant stiffer response than those based on Mori-Tanaka's method at high volume concentration of particles.

Key words particulate polymer composite, viscoelasticity, micromechanics, finite concentration

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Introduction

Mechanical property of polymer composites depends intimately on their microstructure and local phase property. Due to viscoelastic property of the polymeric matrix, polymer composite often presents an overall viscoelastic behavior, especially for particulate composite, such as glass sphere reinforced polymer, plastic binder explosives (PBX)[1–3]. There is an intense study in the past decade to characterize viscoelastic property for polymer composites, particularly to predict the overall viscoelastic behavior from local microstructure and material information of the composite. Based on the correspondence principle between elasticity and linear viscoelasticity[4], Wang and Weng[5] have extended Mori-Tanaka's micromechanical method[6] into the Laplace domain and analyzed the influence of inclusion's shape on the overall viscoelastic behavior of composites. Brinson and Lin[7] further compared the predictions by Mori-Tanaka's method and by finite element computation, and concluded that Mori-Tanaka method can capture the major characteristic for the composite up to a moderate concentration of inclusion. Usually as a rule of thumb, Mori-Tanaka's micromechanical method only gives a good prediction up to 30%[8, 9]. However, there are some polymer composites, whose inclusion concentrations can reach a high value. These composites include polymer foams[10], PBX or particulate composite with a high volume concentration. So it is interesting to propose an analytical method, which can predict the overall viscoelastic behavior for polymer composites at high volume concentration, this is the objective of this paper.

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Since the linear viscoelasticity is solved by a corresponding elastic problem through the correspondence principle, so the key point is to choose an elastic micromechanical model, which can account for high volume concentration of inclusion phase. There are many micromechanical models available for predicting the overall elastic property for composite materials, for example the above-mentioned Mori-Tanaka's method. Self-consistent method[11] and generalized self-consistent method[12] are also popular methods. Other more recent methods incorporate the phase distribution information in their formulations, these methods include Ponte Castañeda and Willis' method[13] (or its equivalent method Kuster-Toksoz model[14, 15]), double inclusion method[16] and IDD method[17]. Their interconnection is discussed recently by Hu and Weng[18, 19]. However, all these methods choose one single pattern (ellipsoidal inclusion or ellipsoidal inclusion with an another ellipsoidal cell of the matrix) and put this single pattern into a reference material to build localization relation[20], the direct short interaction between inclusion phase is only considered in an average sense.

Another approach aims at considering directly two-particle interaction, for example, Molinari and Mouden[21] proposed to solve approximately a multi-particle problem and obtained the corresponding localization relation. Ju and Chen[8] solved approximately two particle interaction problem and obtained the localization relation by introducing statistical probability distribution function. Ma, et al.[9] proposed a generalized Kuster-Toksoz model by considering many pattern interactions. These methods keep the analytical expression and can be applied to a relatively high volume concentration of particles.

In this paper, we will follow the elastic micromechanical method proposed by Ma, et al[9], and utilize the ensemble average method proposed by Ju and Chen[8]. We will propose a method which can predict the overall viscoelastic behavior for particulate polymer composites at high volume concentration. The paper will be arranged as follows: In Section 1, theoretical formulations will be given. First we will give the solution of two-particle interaction in Laplace transformed domain. Then based on this solution, the effective property for composite will be derived in Laplace transformed domain. In Section 2, we will give some numerical examples to illustrate the rate-dependence behavior for particulate polymer composites. Creep curves and stress-strain curves at constant strain rate for glass-bead/ED6 resin composite will be evaluated. The conclusion will be given in Section 3.

1 Theoretical formulation

1.1 Constitutive equation in Laplace transformed domain

In this paper the considered composites are composed of a linear viscoelastic matrix and elastic particles. The matrix and particles are both isotropic materials and the particles are spherical in shape and randomly dispersed in the matrix. So the composite as a whole is also isotropic. In the following discussion, the matrix is referred as the phase 0 and the particle as the phase 1.

For the matrix, the constitutive relation in an integral form can be written as

$$\sigma_{kk}^0 = \int_{-\infty}^{t} 3K_0(t-\tau)\frac{d\varepsilon_{kk}^0(\tau)}{d\tau}\,d\tau, \quad s_{ij}^0 = \int_{-\infty}^{t} 2G_0(t-\tau)\frac{de_{ij}^0(\tau)}{d\tau}\,d\tau, \tag{1a}$$

or,

$$\varepsilon_{kk}^0 = \int_{-\infty}^{t} \frac{I_0(t-\tau)}{3}\frac{d\sigma_{kk}^0(\tau)}{d\tau}\,d\tau, \quad e_{ij}^0 = \int_{-\infty}^{t} \frac{H_0(t-\tau)}{2}\frac{ds_{ij}^0(\tau)}{d\tau}\,d\tau, \tag{1b}$$

where σ_{kk}^0, s_{ij}^0 are respectively deviatoric and spherical parts of the stress σ_{ij}^0; $K_0(t)$, $G_0(t)$ are the bulk and shear relaxation moduli, respectively; ε_{kk}^0, e_{ij}^0 are deviatoric and spherical parts of strain ε_{ij}^0, respectively; $I_0(t)$, $H_0(t)$ are the bulk and shear creep compliances.

The Laplace transformation of a function $f(t)$ is noted by $\tilde{f}(s)$, which is defined by

$$\tilde{f}(s) = \int_0^\infty f(t)\mathrm{e}^{-st}\,dt. \tag{2}$$

Application of Laplace transformation to Eq.(1) leads to

$$\tilde{\sigma}^0_{kk}(s) = 3s\tilde{K}_0(s)\tilde{\varepsilon}^0_{kk}(s), \quad \tilde{s}^0_{ij}(s) = 2s\tilde{G}_0(s)\tilde{e}^0_{ij}(s), \tag{3a}$$

$$\tilde{\varepsilon}^0_{kk}(s) = \frac{s\tilde{I}_0(s)}{3}\tilde{\sigma}^0_{kk}(s), \quad \tilde{e}^0_{ij}(s) = \frac{s\tilde{H}_0(s)}{2}\tilde{s}^0_{ij}(s). \tag{3b}$$

By defining the corresponding bulk and shear moduli of the matrix in the Laplace transformed domain (TD) $\kappa_0^{\mathrm{TD}}(s)$, $\mu_0^{\mathrm{TD}}(s)$, Eq.(3) can be rewritten as[5]

$$\tilde{\sigma}^0_{kk}(s) = 3\kappa_0^{\mathrm{TD}}(s)\tilde{\varepsilon}^0_{kk}(s), \quad \tilde{s}^0_{ij}(s) = 2\mu_0^{\mathrm{TD}}(s)\tilde{e}^0_{ij}(s), \tag{4}$$

and the corresponding bulk and shear moduli in the Laplace transformed domain (TD) are[5]

$$\kappa_0^{\mathrm{TD}}(s) = s\tilde{K}_0(s) = \frac{1}{s\tilde{I}_0(s)}, \quad \mu_0^{\mathrm{TD}}(s) = s\,\tilde{G}_0(s) = \frac{1}{s\tilde{H}_0(s)}. \tag{5}$$

The corresponding Young's modulus and Poisson's ratio in transformed domain are defined as: $E_0^{\mathrm{TD}} = 9\kappa_0^{\mathrm{TD}}\mu_0^{\mathrm{TD}}/(3\kappa_0^{\mathrm{TD}}+\mu_0^{\mathrm{TD}})$, $\nu_0^{\mathrm{TD}} = (3\kappa_0^{\mathrm{TD}}-2\mu_0^{\mathrm{TD}})/(6\kappa_0^{\mathrm{TD}}+2\mu_0^{\mathrm{TD}})$. Application of Laplace transformation to the constitutive equation of elastic particle gives

$$\tilde{\sigma}^1_{kk}(s) = 3\kappa_1\tilde{\varepsilon}^1_{kk}(s), \quad \tilde{s}^1_{ij}(s) = 2\mu_1\tilde{e}^1_{ij}(s), \tag{6}$$

where κ_1, μ_1 are bulk and shear moduli of the particles. So the corresponding bulk and shear moduli in the transformed domain are

$$\kappa_1^{\mathrm{TD}} = \kappa_1, \ \mu_1^{\mathrm{TD}} = \mu_1. \tag{7}$$

With above preparation, we can determine the effective modulus of the composite in the Laplace transformed domain in the following.

1.2 Approximate solutions of two-particle interaction problem in Laplace transformed domain

In this section, we will solve approximately the following problem: two identical spherical particles are embedded into an infinite isotropic matrix. The particles are isotropic elastic materials and the matrix is linear viscoelastic material. The whole is under a remote uniform strain $\boldsymbol{\varepsilon}^0$, as shown in Fig.1. All the analysis will be performed in the Laplace transformed domain.

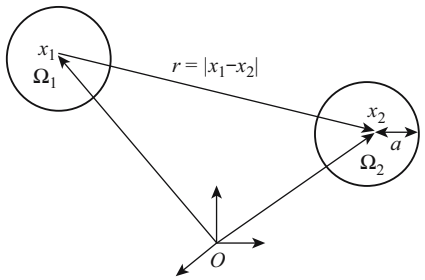

Fig.1 Two-particle interaction problem

According to the method proposed by Zeller and Dederichs[22], the strain in the particle (1) is

$$\tilde{\boldsymbol{\varepsilon}}^{(1)}(\boldsymbol{x}) = \tilde{\boldsymbol{\varepsilon}}^{0} + \int_{\Omega_1} \boldsymbol{\Gamma}^{\mathrm{TD}}(\boldsymbol{x}-\boldsymbol{x}') : \boldsymbol{dL}^{\mathrm{TD}} : \tilde{\boldsymbol{\varepsilon}}^{(1)}(\boldsymbol{x}')\, d\boldsymbol{x}' + \int_{\Omega_2} \boldsymbol{\Gamma}^{\mathrm{TD}}(\boldsymbol{x}-\boldsymbol{x}') : \boldsymbol{dL}^{\mathrm{TD}} : \tilde{\boldsymbol{\varepsilon}}^{(2)}(\boldsymbol{x}')\, d\boldsymbol{x}', \tag{8}$$

where $\boldsymbol{\Gamma}^{\mathrm{TD}}$ is the corresponding tensor of $\boldsymbol{\Gamma}$ tensor in transformed domain. For isotropic materials,

$$\boldsymbol{\Gamma}^{\mathrm{TD}} = \frac{-1}{16\pi\mu_0^{\mathrm{TD}}(1-\nu_0^{\mathrm{TD}})r^3}\boldsymbol{F}(-15, 3\nu_0^{\mathrm{TD}}, 3, 3, -1, 1-2\nu_0^{\mathrm{TD}}), \tag{9}$$

$\boldsymbol{F}(B_m)$ is a four-rank tensor which depends on $(B_1, B_2, B_3, B_4, B_5, B_6)$[8], and

$$\begin{aligned} F_{ijkl}(B_m) =& B_1 n_i n_j n_k n_l + B_2(\delta_{ik}n_j n_l + \delta_{il}n_j n_k + \delta_{jk}n_i n_l + \delta_{jl}n_i n_k) \\ &+ B_3\delta_{ij}n_k n_l + B_4\delta_{kl}n_i n_j + B_5\delta_{ij}\delta_{kl} + B_6(\delta_{ik}\delta_{jk} + \delta_{il}\delta_{jk}), \end{aligned} \tag{10}$$

$\boldsymbol{dL}^{\mathrm{TD}} = \boldsymbol{L}_1^{\mathrm{TD}} - \boldsymbol{L}_0^{\mathrm{TD}}$, $r = |\boldsymbol{x}-\boldsymbol{x}'|$, $\boldsymbol{n} = \frac{\boldsymbol{x}-\boldsymbol{x}'}{r}$. $\boldsymbol{L}_0^{\mathrm{TD}}$, $\boldsymbol{L}_1^{\mathrm{TD}}$ are moduli of the matrix and inclusions respectively.

If we neglect direct particle interaction, according to Eshelby[23], the strain in particles, $\tilde{\boldsymbol{\varepsilon}}_1^0$, can be expressed as

$$\tilde{\boldsymbol{\varepsilon}}_1^0 = \tilde{\boldsymbol{\varepsilon}}^0 - \boldsymbol{P}^{\mathrm{TD}} : \boldsymbol{dL}^{\mathrm{TD}} : \tilde{\boldsymbol{\varepsilon}}_1^0, \tag{11}$$

where $\boldsymbol{P}^{\mathrm{TD}}$ is the corresponding tensor of $\boldsymbol{P}$ tensor in transformed domain and $\boldsymbol{P}^{\mathrm{TD}} = -\int_{\Omega_1}\boldsymbol{\Gamma}^{\mathrm{TD}}(\boldsymbol{x}-\boldsymbol{x}')d\boldsymbol{x}'$. For isotropic materials, $\boldsymbol{P}^{\mathrm{TD}}$ can be expressed by

$$P_{ijkl}^{\mathrm{TD}} = -\frac{1}{30\mu_0^{\mathrm{TD}}(1-\nu_0^{\mathrm{TD}})}\delta_{ij}\delta_{kl} + \frac{4-5\nu_0^{\mathrm{TD}}}{30\mu_0^{\mathrm{TD}}(1-\nu_0^{\mathrm{TD}})}(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}). \tag{12}$$

Subtracting (11) from (8), the strain disturbance in the particle (1) caused by particle (2) can be expressed by

$$\begin{aligned} \tilde{\boldsymbol{d}}^{(1)}(\boldsymbol{x}) =& \Big(\int_{\Omega_2}\boldsymbol{\Gamma}^{\mathrm{TD}}(\boldsymbol{x}-\boldsymbol{x}') : \boldsymbol{dL}^{\mathrm{TD}}\, d\boldsymbol{x}'\Big) : \tilde{\boldsymbol{\varepsilon}}_1^0 + \int_{\Omega_1}\boldsymbol{\Gamma}^{\mathrm{TD}}(\boldsymbol{x}-\boldsymbol{x}') : \boldsymbol{dL}^{\mathrm{TD}} : \tilde{\boldsymbol{d}}^{(1)}(\boldsymbol{x}')\, d\boldsymbol{x}' \\ &+ \int_{\Omega_2}\boldsymbol{\Gamma}^{\mathrm{TD}}(\boldsymbol{x}-\boldsymbol{x}') : \boldsymbol{dL}^{\mathrm{TD}} : \tilde{\boldsymbol{d}}^{(2)}(\boldsymbol{x}')\, d\boldsymbol{x}', \end{aligned} \tag{13}$$

where the strain disturbance is defined as $\tilde{\boldsymbol{d}}^{(i)}(\boldsymbol{x}) = \tilde{\boldsymbol{\varepsilon}}^{(i)}(\boldsymbol{x}) - \tilde{\boldsymbol{\varepsilon}}_1^0$, $i = 1, 2$. Performing volume average in particle (1) to Eq.(13) leads to

$$\begin{aligned} \bar{\tilde{\boldsymbol{d}}}^{(1)} =& \frac{1}{\Omega}\Big[\Big(\int_{\Omega_1}\int_{\Omega_2}\boldsymbol{\Gamma}^{\mathrm{TD}}(\boldsymbol{x}-\boldsymbol{x}')\, d\boldsymbol{x}'d\boldsymbol{x}\Big) : \boldsymbol{dL}^{\mathrm{TD}} : \tilde{\boldsymbol{\varepsilon}}_1^0 \\ &+ \int_{\Omega_1}\int_{\Omega_1}\boldsymbol{\Gamma}^{\mathrm{TD}}(\boldsymbol{x}-\boldsymbol{x}') : \boldsymbol{dL}^{\mathrm{TD}} : \tilde{\boldsymbol{d}}^{(1)}(\boldsymbol{x}')\, d\boldsymbol{x}'d\boldsymbol{x} \\ &+ \int_{\Omega_1}\int_{\Omega_2}\boldsymbol{\Gamma}^{\mathrm{TD}}(\boldsymbol{x}-\boldsymbol{x}') : \boldsymbol{dL}^{\mathrm{TD}} : \tilde{\boldsymbol{d}}^{(2)}(\boldsymbol{x}')\, d\boldsymbol{x}'d\boldsymbol{x}\Big], \end{aligned} \tag{14}$$

where $\bar{\tilde{\boldsymbol{d}}}^{(i)}$ is the average strain disturbance. It is defined by $\bar{\tilde{\boldsymbol{d}}}^{(i)} = \frac{1}{\Omega}\int_{\Omega_i}\tilde{\boldsymbol{d}}^{(i)}(\boldsymbol{x})\, d\boldsymbol{x}$. Assuming the strain in a particle is uniform, it has

$$\bar{\tilde{\boldsymbol{d}}}^{(1)} \approx -\boldsymbol{P}_2^{\mathrm{TD}} : \boldsymbol{dL}^{\mathrm{TD}} : \tilde{\boldsymbol{\varepsilon}}_1^0 - \boldsymbol{P}^{\mathrm{TD}} : \boldsymbol{dL}^{\mathrm{TD}} : \bar{\tilde{\boldsymbol{d}}}^{(1)} - \boldsymbol{P}_2^{\mathrm{TD}} : \boldsymbol{dL}^{\mathrm{TD}} : \bar{\tilde{\boldsymbol{d}}}^{(2)}, \tag{15}$$

$$\boldsymbol{P}_2^{\mathrm{TD}}(\boldsymbol{x}_1-\boldsymbol{x}_2) = -\frac{1}{\Omega}\int_{\Omega_1}\int_{\Omega_2}\boldsymbol{\Gamma}^{\mathrm{TD}}(\boldsymbol{x}-\boldsymbol{x}')\, d\boldsymbol{x}'d\boldsymbol{x} = \frac{1}{60\mu_0^{\mathrm{TD}}(1-\nu_0^{\mathrm{TD}})}(\rho^3\boldsymbol{H}^0 + 2\rho^5\boldsymbol{H}^2), \tag{16}$$

$\boldsymbol{H}^0(\boldsymbol{x}_1-\boldsymbol{x}_2)=5\boldsymbol{F}(-15,3\nu_0^{\rm TD},3,3,-1,1-2\nu_0^{\rm TD})$, $\boldsymbol{H}^2(\boldsymbol{x}_1-\boldsymbol{x}_2)=3\boldsymbol{F}(35,-5,-5,-5,1,1)$, $\rho=\frac{a}{r_0}$, $r_0=|\boldsymbol{x}_1-\boldsymbol{x}_2|$, $\boldsymbol{x}_i$ is the central point of Ω_i, a is radius of the particle and Ω is its volume.

By applying the same method for the particle (2), we can get $\bar{\tilde{\boldsymbol{d}}}^{(1)}=\bar{\tilde{\boldsymbol{d}}}^{(2)}=\bar{\tilde{\boldsymbol{d}}}_1$. So it can be followed that

$$\begin{aligned}\bar{\tilde{\boldsymbol{d}}}_1&=-[\boldsymbol{I}+\boldsymbol{P}^{\rm TD}:\boldsymbol{dL}^{\rm TD}+\boldsymbol{P}_2^{\rm TD}:\boldsymbol{dL}^{\rm TD}]^{-1}:\boldsymbol{P}_2^{\rm TD}:\boldsymbol{dL}^{\rm TD}:\tilde{\boldsymbol{\varepsilon}}_1^0\\&=-\boldsymbol{L}_0^{\rm TD}:(\boldsymbol{L}_0^{\rm TD}+\boldsymbol{P}^{\rm TD}:\boldsymbol{L}_0^{\rm TD}:\boldsymbol{dL}^{\rm TD})^{-1}:\boldsymbol{R}^{-1}:\boldsymbol{P}_2^{\rm TD}:\boldsymbol{dL}^{\rm TD}:\tilde{\boldsymbol{\varepsilon}}_1^0,\end{aligned}\tag{17}$$

and $\boldsymbol{R}=(\boldsymbol{I}+\boldsymbol{P}_2^{\rm TD}:\boldsymbol{L}_0^{\rm TD}:\boldsymbol{Q})$, $\boldsymbol{Q}=\boldsymbol{dL}^{\rm TD}:(\boldsymbol{L}_0^{\rm TD}+\boldsymbol{P}^{\rm TD}:\boldsymbol{L}_0^{\rm TD}:\boldsymbol{dL}^{\rm TD})^{-1}$. Let $\boldsymbol{Q}=\boldsymbol{F}\{0,0,0,0,\alpha,\beta\}$, then,

$$\alpha=\frac{3(1-\nu_0^{\rm TD})(1-5\nu_0^{\rm TD})d\kappa^{\rm TD}d\mu^{\rm TD}+15(1-\nu_0^{\rm TD})^2(d\kappa^{\rm TD}\mu_0^{\rm TD}-d\mu^{\rm TD}\kappa_0^{\rm TD})}{[(1+\nu_0^{\rm TD})d\kappa^{\rm TD}+3(1-\nu_0^{\rm TD})\kappa_0^{\rm TD}][15(1-\nu_0^{\rm TD})\mu_0^{\rm TD}+2(4-5\nu_0^{\rm TD})d\mu^{\rm TD}]},\tag{18a}$$

$$\beta=\frac{15(1-\nu_0^{\rm TD})d\mu^{\rm TD}}{30(1-\nu_0^{\rm TD})\mu_0^{\rm TD}+4(4-5\nu_0^{\rm TD})d\mu^{\rm TD}},\tag{18b}$$

and $d\kappa^{\rm TD}=\kappa_1^{\rm TD}-\kappa_0^{\rm TD}$, $d\mu^{\rm TD}=\mu_1^{\rm TD}-\mu_0^{\rm TD}$. The Taylor expansion of $\boldsymbol{R}^{-1}$ in regard of $\rho=0$ can be written as

$$\boldsymbol{R}^{-1}=\boldsymbol{I}+\rho^3\boldsymbol{M}+\cdots,\tag{19}$$

$$\begin{aligned}\boldsymbol{M}(\boldsymbol{x}_1-\boldsymbol{x}_2)=-\frac{\beta}{3(1-\nu_0^{\rm TD})}\{&-15,3\nu_0^{\rm TD},\ 3,\ 3(1-2\nu_0^{\rm TD})-\frac{3(1+\nu_0^{\rm TD})\alpha}{\beta},\\&-1+2\nu_0^{\rm TD}+\frac{(1+\nu_0^{\rm TD})\alpha}{\beta},\quad 1-2\nu_0^{\rm TD}\}.\end{aligned}\tag{20}$$

The formulations of $\boldsymbol{F}^{-1}$ and $\boldsymbol{F}(A_m):\boldsymbol{F}(B_m)$ can be found in appendix of Ref.[8].

With the help of expressions of R^{-1} and $P_2^{\rm TD}$, the average strain disturbance can be rewritten as

$$\bar{\tilde{\boldsymbol{d}}}(\boldsymbol{x}_1-\boldsymbol{x}_2)=-\frac{\boldsymbol{L}_0^{\rm TD}}{60\mu_0^{\rm TD}(1-\nu_0^{\rm TD})}:(\boldsymbol{L}_0^{\rm TD}+\boldsymbol{P}^{\rm TD}:\boldsymbol{L}_0^{\rm TD}:\boldsymbol{dL}^{\rm TD})^{-1}:\boldsymbol{A}:\boldsymbol{dL}^{\rm TD}:\tilde{\boldsymbol{\varepsilon}}_1^0,\tag{21}$$

$$\boldsymbol{A}=(\rho^3\boldsymbol{H}^0+2\rho^5\boldsymbol{H}^2)+\rho^6\boldsymbol{M}:\boldsymbol{H}^0+o(\rho^8)\approx(\rho^3\boldsymbol{H}^0+2\rho^5\boldsymbol{H}^2)+\rho^6\boldsymbol{M}:\boldsymbol{H}^0.\tag{22}$$

1.3 Ensemble-average solution

For a many-particle problem, according to the ensemble-average method proposed by Ju and Chen[8], the strain disturbance in particle (1) caused by all other particles can be solved by integrating $\bar{\tilde{\boldsymbol{d}}}_1$ over all possible position of other particles when particle (1) is fitted at $\boldsymbol{x}_1$. It can be expressed as[8]

$$\langle\bar{\tilde{\boldsymbol{d}}}_1\rangle(\boldsymbol{x}_1)=\int_{V-\Omega_1}\bar{\tilde{\boldsymbol{d}}}_1(\boldsymbol{x}_1-\boldsymbol{x}_2)P(\boldsymbol{x}_2/\boldsymbol{x}_1)\,d\boldsymbol{x}_2,\tag{23}$$

where $\langle\bar{\tilde{\boldsymbol{d}}}_1\rangle$ is ensemble-average strain disturbance , and the angled bracket means the ensemble-average. $P(\boldsymbol{x}_2/\boldsymbol{x}_1)$ is conditional probability distribution function, which means the probability of finding the second particle centered at $\boldsymbol{x}_2$ for a given particle centered at $\boldsymbol{x}_1$. If we further assume that $P(\boldsymbol{x}_2/\boldsymbol{x}_1)$ is statistically isotropic, we have

$$P(\boldsymbol{x}_2/\boldsymbol{x}_1)=\begin{cases}\frac{N}{V}g(r), & r\geq 2a;\\ 0, & \text{otherwise},\end{cases}\tag{24}$$

where N/V is the number density of particles in the composite.

In this paper, two isotropic distribution functions are considered: uniform distribution function ($g(r) = 1$) and Percus-Yevick (PY) radial distribution function[24, 25]. For composites with randomly dispersed non-overlapping spheres, PY distribution function is considered to be the most reliable function. Figure 2 shows PY radial distribution function for volume fractions, $\varphi = 0.05, 0.25, 0.5$.

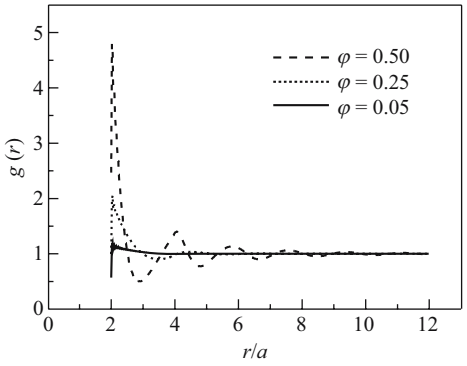

Fig.2 PY radial distribution function

With help of Eqs.(21) and (24), Eq.(23) can be rewritten as

$$\langle\bar{\tilde{\boldsymbol{d}}}_1\rangle(\boldsymbol{x}_1) = -\frac{N}{V}\frac{\boldsymbol{L}_0^{\mathrm{TD}}}{60\mu_0^{\mathrm{TD}}(1-\nu_0^{\mathrm{TD}})} : (\boldsymbol{L}_0^{\mathrm{TD}} + \boldsymbol{P}^{\mathrm{TD}} : \boldsymbol{L}_0^{\mathrm{TD}} : \boldsymbol{dL}^{\mathrm{TD}})^{-1} : \boldsymbol{A}_1 : \boldsymbol{dL}^{\mathrm{TD}} : \tilde{\boldsymbol{\varepsilon}}_1^0, \tag{25}$$

$$\begin{aligned}\boldsymbol{A}_1 = &\int_{2a}^{\infty} \rho^3 g(r) \int_{\Xi} \boldsymbol{H}^0(\boldsymbol{n})\, d\,\Xi dr + 2\int_{2a}^{\infty} \rho^5 g(r) \int_{\Xi} \boldsymbol{H}^2(\boldsymbol{n})\, d\,\Xi dr \\ &+ \int_{2a}^{\infty} \rho^6 g(r) \int_{\Xi} \boldsymbol{M} : \boldsymbol{H}^0(\boldsymbol{n})\, d\,\Xi dr,\end{aligned} \tag{26}$$

where Ξ denotes spherical surface of radius r, and $\boldsymbol{n}$ is normal vector of a point on Ξ.

It can be easily proved that the following identities are valid[8]:

$$\int_{\Xi} n_i n_j\, d\,\Xi = \frac{4\pi r^2}{3}\delta_{ij}, \quad \int_{\Xi} n_i n_j n_k n_l\, d\,\Xi = \frac{4\pi r^2}{15}(\delta_{ij}\delta_{kl} + \delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}).$$

Then we can obtain equation:

$$\int_{\Xi} F_{ijkl}(B_m)\, d\Xi = 4\pi[\eta_1\delta_{ij}\delta_{kl} + \eta_2(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})]r^2,$$

where $\eta_1 = \frac{B_1}{15} + \frac{B_3+B_4}{3} + B_5$, $\quad \eta_2 = \frac{B_1}{15} + \frac{2B_2}{3} + B_6$.

Thus we can also get that,

$$\int_{\Xi} \boldsymbol{H}^0 d\Xi = 0, \quad \int_{\Xi} \boldsymbol{H}^2 d\Xi = 0.$$

Therefore $\boldsymbol{A}_1$ can be simplified as

$$\boldsymbol{A}_1 = \int_{2a}^{\infty} \rho^6 g(r) \int_{\Xi} \boldsymbol{M} : \boldsymbol{H}^0(\boldsymbol{n}) d\Xi dr. \tag{27}$$

It can be observed that $\boldsymbol{A}_1$ has the form of $\boldsymbol{F}(0, 0, 0, 0, B_5, B_6)$. So the following identity is valid, $\boldsymbol{A}_1 : \boldsymbol{dL}^{\mathrm{TD}} = \boldsymbol{dL}^{\mathrm{TD}} : \boldsymbol{A}_1$. Then the ensemble-average strain disturbance can be simplified as

$$\langle\bar{\tilde{\boldsymbol{d}}}_1\rangle(\boldsymbol{x}_1) = -\frac{N}{V}\frac{\boldsymbol{L}_0^{\mathrm{TD}}}{60\mu_0^{\mathrm{TD}}(1-\nu_0^{\mathrm{TD}})} : \boldsymbol{Q} : \boldsymbol{A}_1 : \tilde{\boldsymbol{\varepsilon}}_1^0. \tag{28}$$

Finally the ensemble-average strain disturbance can be written as

$$\langle\bar{\tilde{\boldsymbol{d}}}_1\rangle(\boldsymbol{x}_1) = Y(g)\boldsymbol{W} : \tilde{\boldsymbol{\varepsilon}}_1^0, \tag{29}$$

and

$$Y(g) = \int_{2a}^{\infty} \frac{a^3}{r^4} g(r)dr = \int_0^{1/2} \rho^2 g(\rho)d\rho, \quad W_{ijkl} = \xi_1\delta_{ij}\delta_{kl} + \xi_2(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk}),$$

$$\xi_1 = \frac{2\beta\varphi}{15(1-\nu_0^{\mathrm{TD}})^2}\{8\alpha[1-\nu_0^{\mathrm{TD}} - 2(\nu_0^{\mathrm{TD}})^2] - 4\beta[2 - 2\nu_0^{\mathrm{TD}} + 5(\nu_0^{\mathrm{TD}})^2]\}, \tag{30a}$$

$$\xi_2 = \frac{2\beta\varphi}{15(1-\nu_0^{\mathrm{TD}})^2}\{3\alpha[1-\nu_0^{\mathrm{TD}} - 2(\nu_0^{\mathrm{TD}})^2] + 2\beta[11 - 11\nu_0^{\mathrm{TD}} + 5(\nu_0^{\mathrm{TD}})^2]\}, \tag{30b}$$

where φ is volume fraction of particles.

According to definition of the disturbance strain, the ensemble-average strain in particles can be expressed as

$$\langle\bar{\tilde{\boldsymbol{\varepsilon}}}_1\rangle = (\boldsymbol{I} + Y(g)\boldsymbol{W}) : \tilde{\boldsymbol{\varepsilon}}_1^0 = \boldsymbol{K}^{\mathrm{TD}} : \tilde{\boldsymbol{\varepsilon}}_1^0, \tag{31}$$

and $K_{ijkl}^{\mathrm{TD}} = \gamma_1\delta_{ij}\delta_{kl} + \gamma_2(\delta_{ik}\delta_{jl} + \delta_{il}\delta_{jk})$, $\gamma_1 = \xi_1 Y(g)$, $\gamma_2 = 1/2 + \xi_2 Y(g)$.

1.4 Effective modulus of composite in Laplace transformed domain

In order to build the effective modulus for the composite material, we will follow the idea proposed by Kuster and Togsoz (see Ref.[9] for more details). The idea can be explained as follows: we put a composite particle into the matrix material under the same remote load $\boldsymbol{\varepsilon}^0$ as previously (see Fig.3). The form of the composite particle characterizes the information of particle distribution. In this paper it is assumed to be spherical in order to characterize the isotropic distribution.

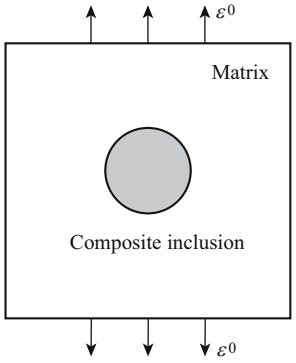

Fig.3 Kuster-Togsoz model

Assuming that the composite is also an isotropic linear viscoelastic material, so its constitutive equation in Laplace transformed domain (TD) can be also written as

$$\tilde{\sigma}_{kk}^{\mathrm{c}}(s) = 3\kappa_{\mathrm{c}}^{\mathrm{TD}}(s)\tilde{\varepsilon}_{kk}^{\mathrm{c}}(s), \quad \tilde{s}_{ij}^{\mathrm{c}}(s) = 2\mu_{\mathrm{c}}^{\mathrm{TD}}(s)\tilde{e}_{ij}^{\mathrm{c}}(s), \tag{32}$$

where "c" is referred as composite. According to Eshelby[23], the strain in the composite particle can be expressed by

$$\tilde{\boldsymbol{\varepsilon}}_{\mathrm{c}} = [\boldsymbol{I} + \boldsymbol{P}^{\mathrm{TD}} : (\boldsymbol{L}_{\mathrm{c}}^{\mathrm{TD}} - \boldsymbol{L}_0^{\mathrm{TD}})]^{-1} : \tilde{\boldsymbol{\varepsilon}}^0, \tag{33}$$

where $\boldsymbol{L}_{\mathrm{c}}^{\mathrm{TD}}$ is the composite modulus in Laplace transformed domain.

With help of Eqs.(11), (31) and (33), the localization relation can be finally written as

$$\langle\bar{\tilde{\boldsymbol{\varepsilon}}}_1\rangle = \boldsymbol{K}^{\mathrm{TD}} : (\boldsymbol{I} + \boldsymbol{P}^{\mathrm{TD}} : \boldsymbol{dL}^{\mathrm{TD}})^{-1} : [\boldsymbol{I} + \boldsymbol{P}^{\mathrm{TD}} : (\boldsymbol{L}_{\mathrm{c}}^{\mathrm{TD}} - \boldsymbol{L}_0^{\mathrm{TD}})] : \tilde{\boldsymbol{\varepsilon}}_{\mathrm{c}}. \tag{34}$$

So the final effective modulus of the composite in Laplace transformed domain is given by

$$\boldsymbol{L}_{\mathrm{c}}^{\mathrm{TD}} = \boldsymbol{L}_0^{\mathrm{TD}} : [\boldsymbol{I} + \varphi((\boldsymbol{K}^{\mathrm{TD}} : \boldsymbol{Q})^{-1} - \varphi\boldsymbol{P}^{\mathrm{TD}} : \boldsymbol{L}_0^{\mathrm{TD}})^{-1}]. \tag{35}$$

Then, the corresponding effective bulk modulus and shear modulus can be evaluated as

$$\frac{\kappa_{\mathrm{c}}^{\mathrm{TD}}}{\kappa_0^{\mathrm{TD}}} = \frac{3(1-\nu_0^{\mathrm{TD}}) + 2\varphi(1-2\nu_0^{\mathrm{TD}})(3\gamma_1 + 2\gamma_2)(3\alpha + 2\beta)}{3(1-\nu_0^{\mathrm{TD}}) - \varphi(1+\nu_0^{\mathrm{TD}})(3\gamma_1 + 2\gamma_2)(3\alpha + 2\beta)}, \tag{36a}$$

$$\frac{\mu_{\mathrm{c}}^{\mathrm{TD}}}{\mu_0^{\mathrm{TD}}} = \frac{15(1-\nu_0^{\mathrm{TD}}) + 4\varphi(7-5\nu_0^{\mathrm{TD}})\beta\gamma_2}{15(1-\nu_0^{\mathrm{TD}}) - 8\varphi(4-5\nu_0^{\mathrm{TD}})\beta\gamma_2}. \tag{36b}$$

The relaxation moduli and creep compliances of the composite can be obtained by applying the inverse of Laplace transformation, that is,

$$K_{\rm c}(t)=L^{-1}\left(\frac{\kappa_{\rm c}^{\rm TD}(s)}{s}\right),\quad G_{\rm c}(t)=L^{-1}\left(\frac{\mu_{\rm c}^{\rm TD}(s)}{s}\right),\tag{37}$$

where L^{-1} is the inverse of the Laplace transformation.

2 Numerical application

In this section, we will consider ED-6 resin-matrix composite with randomly dispersed glass beads. Wang and Weng[5] used four-parameter model to approach creep experimental data of ED-6 resin obtained by Skudra and Auzukalns[26] and determined the four constants of the creep compliance $J(t)=\frac{1}{E_{\rm M}}+\frac{t}{\eta_{\rm M}}+\frac{1}{E_{\rm v}}(1-{\rm e}^{-\frac{E_{\rm v}}{\eta_{\rm v}}t})$ as follows:

$$\begin{aligned}&E_{\rm M}=3.27\ {\rm GPa},\quad E_{\rm v}=1.8\ {\rm GPa},\\&\eta_{\rm M}=8000\ {\rm GPa\cdot h},\quad \eta_{\rm v}=300\ {\rm GPa\cdot h}.\end{aligned}\tag{38}$$

The fitting curve is illustrated in Fig.4.

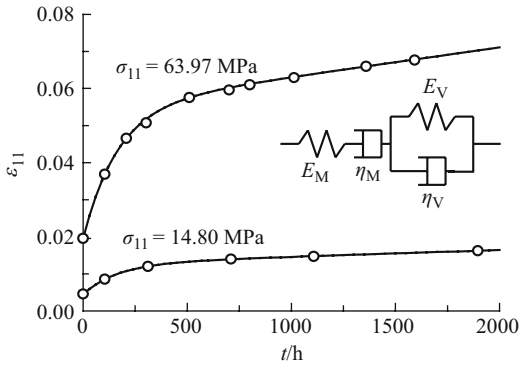

Fig.4 Creep curves of ED-6 resin by four-parameter model

In this paper we assume Poisson's ratio of the matrix is a constant, namely $\nu_0^{\rm TD}=\nu_0=0.38$. The bulk and shear creep compliance of the matrix can be obtained by the following equations: $I(t)=3(1-2\nu_0)J(t)$, $H(t)=2(1+\nu_0)J(t)$. With the help of Eq.(5), the corresponding bulk and shear moduli of the matrix in transformed domain can be expressed as

$$\kappa_0^{\rm TD}(s)=\frac{E_{\rm M}\eta_{\rm M}(E_{\rm v}+\eta_{\rm v}s)s}{3(1-2\nu_0)(E_{\rm M}E_{\rm v}+s(E_{\rm M}\eta_{\rm M}+E_{\rm M}\eta_{\rm v}+E_{\rm v}\eta_{\rm M})+s^2\eta_{\rm M}\eta_{\rm v})},\tag{39a}$$

$$\mu_0^{\rm TD}(s)=\frac{E_{\rm M}\eta_{\rm M}(E_{\rm v}+\eta_{\rm v}s)s}{2(1+\nu_0)(E_{\rm M}E_{\rm v}+s(E_{\rm M}\eta_{\rm M}+E_{\rm M}\eta_{\rm v}+E_{\rm v}\eta_{\rm M})+s^2\eta_{\rm M}\eta_{\rm v})}.\tag{39b}$$

Glass bead can be considered as an elastic material at room temperature and its elastic constants are

$$\kappa_1^{\rm TD}=\kappa_1=39.43\ {\rm GPa},\quad \mu_1^{\rm TD}=\mu_1=28.35\ {\rm GPa}.\tag{40}$$

Having known the material constants of the components, we can evaluate the effective shear and bulk moduli of the composite with Eqs.(36)–(37). In this paper the computing results with uniform distribution function (uniform d.f.) and PY radial distribution function (PY r.d.f.)

are compared with those based on Mori-Tanaka's method. Figures 5(a) and 5(b) show the creep curves under hydrostatic and shear loading for $\varphi = 0.5$. We find that under a hydrostatic loading the response predicted by the proposed method are very close to those predicted by MT method. However under shear loading there is a significant difference between the two methods: MT method predicts the softest response (largest compliance), and the proposed method with PY distribution gives the stiffest one. This is due to the particle-interaction effect, considered by the proposed method, and not considered by MT method. We can also seen that the prediction on creep compliance with PY distribution is smaller than that predicted by uniform distribution. This is because PY distribution function estimates a higher probability of the second particle located in the neighbourhood of the first particle than the uniform distribution. The same trend is also observed for $\varphi = 0.25$, which is shown in Fig.6. By comparing the results shown in Fig.5 and Fig.6, it is found that responses predicted by the two methods tend to be similar as the volume fraction decreases.

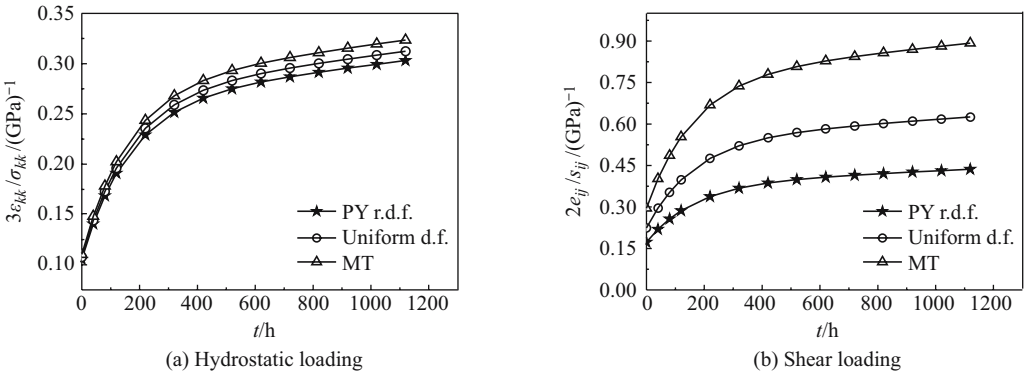

Fig.5 Creep compliance of the composite predicted with different methods ($\varphi = 0.5$)

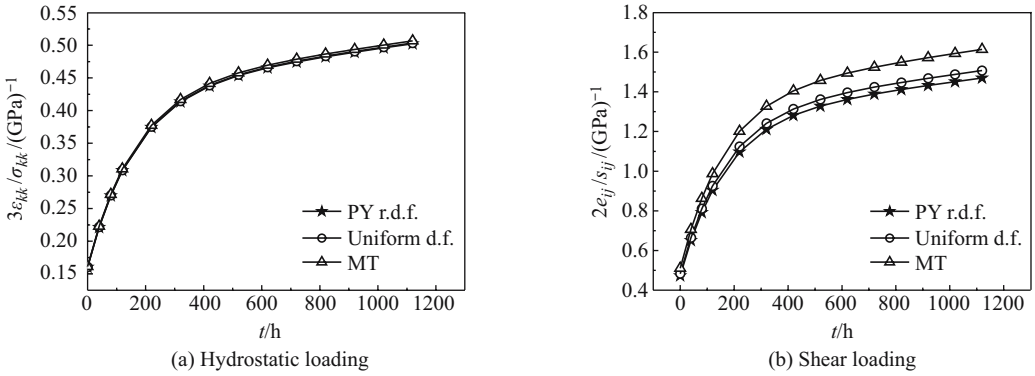

Fig.6 Creep compliance of the composite predicted with different methods ($\varphi = 0.25$)

The stress-strain curves for the composite at a constant strain-rate are illustrated in Fig.7. As expected, the proposed method with PY distribution predicts a stiffer response than those based on Mori-Tanaka's method, especially for high volume concentration.

The influence of strain rate on stress-strain curves is shown in Figs.8(a) and 8(b) for hydrostatic and shear loading, respectively. It can be seen that the strain rate has an important influence on the overall stress and strain relation for viscoleastic composites.

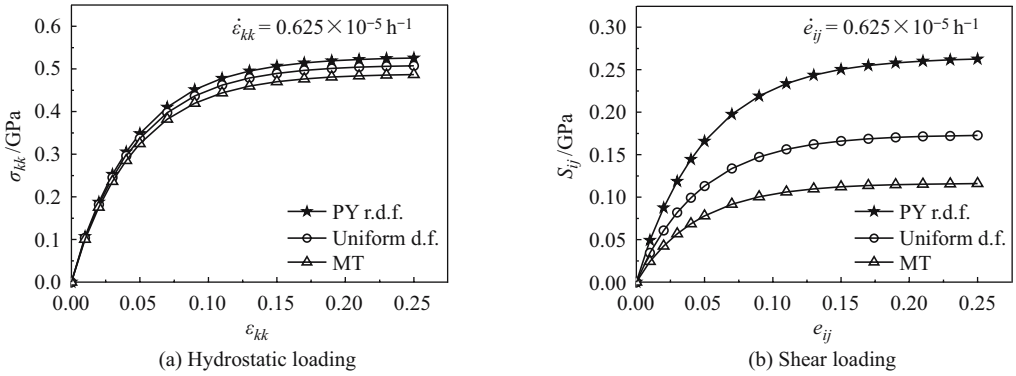

Fig.7 σ-ε curves at constant strain rate predicted by different methods ($\varphi = 0.5$)

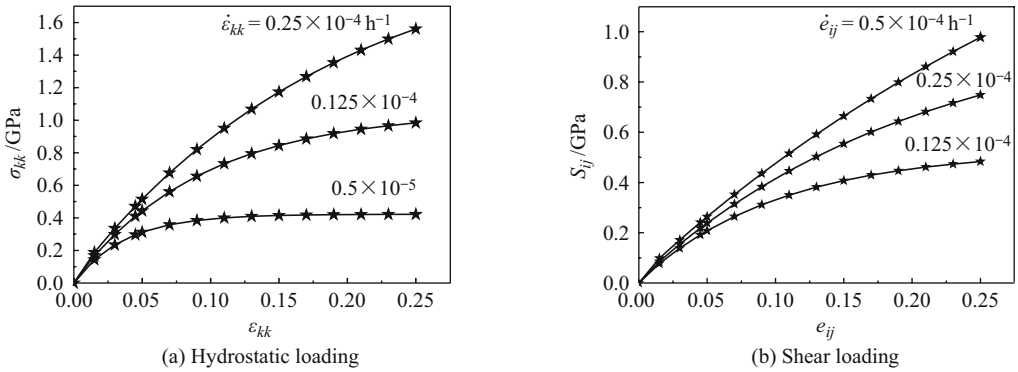

Fig.8 Influence of strain rate on σ-ε curves predicted by the present method with PY distribution ($\varphi = 0.5$)

3 Conclusions

A new micromechanical method is proposed to predict effective viscoelastic property for particulate polymer composites with finite concentration of particles. The method is based on a generalized KT model and the approximate solution of two-particle interaction. By considering the direct interaction between particles, the proposed method can be applied for composites with high volume concentration. The numerical results show that:

(1) A significant difference is observed between the proposed method and Mori-Tanaka's method for high volume concentration of particles.

(2) Under hydrostatic loading the two methods predict similar response. However under shear loading the proposed method predict a significantly stiffer response than that based on MT method.

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