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On Using Micromechanical Models to Describe Dynamic Mechanical Behavior of Asphalt Mastics

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Introduction

Micromechanical modeling has been receiving a great deal of attention recently [1- 4] for interpretation of the mechanical behavior of polymer composites. The models fall in three basic categories: (1) those that are based on phenomenological macroscopic analyses [5, 6] and extended through the correspondence principle [7], (2) those that are based on variational principles [8, 9], and (3) those that are based on the generalized self-consistent schemes [10-12]. The first analytical formulation of the generalized self-consistent scheme for two-phase materials was done by Christensen and Lo [13] through a 3-phase model described by considering a spherical inclusion surrounded by a matrix shell that is embedded in an effective equivalent infinite medium. Extensions to materials with more than two phases were later proposed from the concept of multi-layered inclusions, such as, 4-phase model of Maurer [14] as well as Alberola and Mele [2] and (n+1)-phase model proposed by Herve and Zaoui [15]. Application of the 3-phase model in two successive steps was found by Shaterzadeh et al. [1] to give better prediction than the 4-phase formulation.

Buttlar and Roque [16, 17] extended the concepts of micromechanical modeling from polymer composites to asphalt mixtures and mastics. While none of the available models was found suitable for predicting asphalt mixture properties, it was concluded [16, 17] that micromechanical models had a potential use in predicting asphalt mastic properties. With a view to check these conclusions, Buttlar et al [18] made an attempt at understanding asphalt mastic behavior through micromechanics. An equivalent rigid-layer modeling concept was introduced and an attempt was made to explain the findings. However, there were several questions raised [19] regarding the work [18], some of which are addressed in this paper.

The present work goes through a systematic derivation to get the correct simplified form of the equation based on micromechanical modeling. It then evaluates the efficiency of the model in correctly predicting the stiffness ratio of asphalt mastics. In order to overcome the deficiencies of the model at higher volume fractions of fillers, the concepts of percolation theory are made use and a modified version of the model is proposed. Comparison of this modified version with experimental data is shown to yield realistic trends. The merits and limitations of the model are discussed in detail.

Theoretical Development

The micromechanical model that is chosen for describing the dynamic mechanical behavior of asphalt mastics is the Christensen and Lo model [13]. It is based on a "generalized self-consistent scheme", at times referred to as the GSCS, in which the system is considered as a composite having a spherical inclusion surrounded by a matrix shell within a homogeneous medium as shown in Figure 1. The asphalt mastic is visualized to fit this 3-phase model description and the constitutive equation for this model as given by Christensen and Lo [13] is written in a form applicable to asphalt mastics as follows:

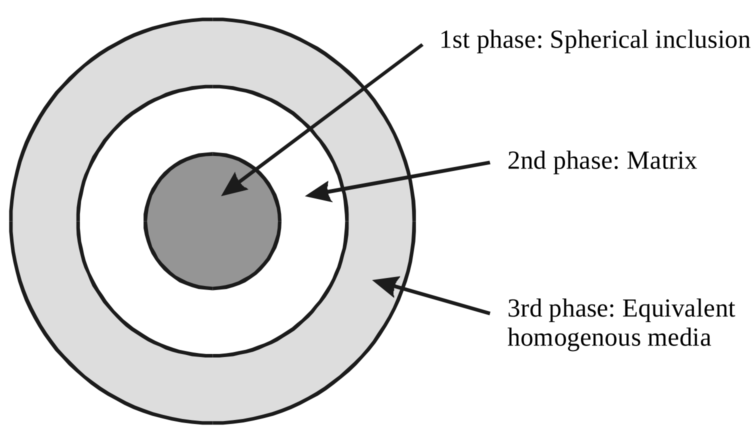


Figure 1. Three-phase model description of the asphalt mastic behavior befitting the "generalized self-consistent scheme".

$$A\left(\frac{G_c}{G_m}\right)^2 + B\left(\frac{G_c}{G_m}\right) + C = 0 \tag{1}$$

The expressions for A, B and C are

$$A = 8\left(\left(\frac{G_f}{G_m}\right) - 1\right)(4 - 5\nu_m)\eta_1\phi_f^{\left(\frac{10}{3}\right)} - 2\left(63\left(\left(\frac{G_f}{G_m}\right) - 1\right)\eta_2 + 2\eta_1\eta_3\right)\phi_f^{\left(\frac{7}{3}\right)}$$
$$+252\left(\left(\frac{G_f}{G_m}\right) - 1\right)\eta_2\phi_f^{\left(\frac{5}{3}\right)} - 50\left(\left(\frac{G_f}{G_m}\right) - 1\right)\left(7 - 12\nu_m + 8\nu_m^2\right)\eta_2\phi_f + 4(7 - 10\nu_m)\eta_2\eta_3 \tag{2a}$$

$$B = -4\left(\left(\frac{G_f}{G_m}\right) - 1\right)(1 - 5\nu_m)\eta_1\phi_f^{\left(\frac{10}{3}\right)} + 4\left(63\left(\left(\frac{G_f}{G_m}\right) - 1\right)\eta_2 + 2\eta_1\eta_3\right)\phi_f^{\left(\frac{7}{3}\right)}$$
$$-504\left(\left(\frac{G_f}{G_m}\right) - 1\right)\eta_2\phi_f^{\left(\frac{5}{3}\right)} + 150\left(\left(\frac{G_f}{G_m}\right) - 1\right)(3 - \nu_m)\nu_m\eta_2\phi_f + 3(15\nu_m - 7)\eta_2\eta_3 \tag{2b}$$

$$C = 4\left(\left(\frac{G_f}{G_m}\right) - 1\right)(5\nu_m - 7)\eta_1\phi_f^{\left(\frac{10}{3}\right)} - 2\left(63\left(\left(\frac{G_f}{G_m}\right) - 1\right)\eta_2 + 2\eta_1\eta_3\right)\phi_f^{\left(\frac{7}{3}\right)}$$
$$+252\left(\left(\frac{G_f}{G_m}\right) - 1\right)\eta_2\phi_f^{\left(\frac{5}{3}\right)} + 25\left(\left(\frac{G_f}{G_m}\right) - 1\right)\left(\nu_m^2 - 7\right)\eta_2\phi_f - (7 + 5\nu_m)\eta_2\eta_3 \tag{2c}$$

where

$$\eta_1 = \left(\left(\frac{G_f}{G_m}\right) - 1\right)(49 - 50\nu_f\nu_m) + 35\left(\left(\frac{G_f}{G_m}\right)(\nu_f - 2\nu_m) + (2\nu_f - \nu_m)\right) \tag{3a}$$

$$\eta_2 = 5\nu_f\left(\left(\frac{G_f}{G_m}\right) - 8\right) + 7\left(\left(\frac{G_f}{G_m}\right) + 4\right) \tag{3b}$$

$$\eta_3 = \left(\left(\frac{G_f}{G_m}\right)(8 - 10\nu_m) + (7 - 5\nu_m)\right) \tag{3c}$$

G corresponds to the shear modulus, ν the Poisson's ratio, ϕ the volume fraction while the subscripts c, m and f denote the composite, matrix and filler, respectively. The composite in the present case is the mastic while the matrix is the asphalt and the fine aggregates are the fillers.

The GSCS in the form given by equations (1) – (3) is cumbersome to use routinely and hence it is natural to look for means of simplification. Buttlar et al [18] went through a least-squares nonlinear regression and suggested an exponential expression as:

$$\left(\frac{G_c}{G_m}\right) = a\exp\left(bG_m\right) \tag{4}$$

where $a = 25.083\ \phi_f^3 - 10.154\ \phi_f^2 + 4.8767\ \phi_f + 0.831$;

and $b = (-1.28\times10^{-3})\ \phi_f^3 - (3.37\times10^{-4})\ \phi_f^2 + (3.08\times10^{-4})\ \phi_f + (6.53\times10^{-6})$.

It is seen that the stiffness ratio is shown to be dependent on the stiffness of the matrix G_m. There are empirical relations in the literature [20-22], which suggest that this might be the case. A closer look reveals that they [20-22] are all considering the hypothetical case of the bitumen attaining a stiffness value equal to that of the aggregate. While at low temperatures, the stiffness of the bitumen will come closer to the stiffness of the filler, it can never approach the stiffness of the filler. As a matter of the fact, the stiffness of the binder and the filler will be two orders apart. This can be effectively used to simplify equations (1) - (3).

In the age of computers, it is not very difficult to compute the shear modulus from the Christensen and Lo [13] equations in their original form. However, a simplification of the equations is often useful in order to actiually see which terms dominate and which terms do not, thereby giving a better perspective and insight into how the model operates and predicts. Hence a simplification of the Christensen and Lo [13] model is sought.

Simplification of the Christensen and Lo model

The simplification process essentially involves the use of an order of magnitude analysis. All terms in the expressions that are insignificant, in comparison to others with much larger magnitude, are identified so that they could be ignored. In the case of asphalt mastics, it can be safely assumed that $G_f >> G_m$, or in other words, $G_f/G_m >> 1$ or even 10. Thus, the following equalities would approximately hold without much error, also knowing that $0 < \nu_f , \nu_m < 1$.

$$\left(\left(\frac{G_f}{G_m}\right) - 1\right) \approx \left(\frac{G_f}{G_m}\right) \tag{5a}$$

$$\left(\left(\frac{G_f}{G_m}\right) - 8\right) \approx \left(\frac{G_f}{G_m}\right) \tag{5b}$$

$$\left(\left(\frac{G_f}{G_m}\right) + 4\right) \approx \left(\frac{G_f}{G_m}\right) \tag{5c}$$

$$\left(\left(\frac{G_f}{G_m}\right)\left(\nu_f - 2\nu_m\right) + \left(2\nu_f - \nu_m\right)\right) \approx \left(\frac{G_f}{G_m}\right)\left(\nu_f - 2\nu_m\right) \tag{5d}$$

$$\left(\left(\frac{G_f}{G_m}\right)\left(8 - 10\nu_m\right) + \left(7 - 5\nu_m\right)\right) \approx \left(\frac{G_f}{G_m}\right)\left(8 - 10\nu_m\right) \tag{5e}$$

Using equations (5a) – (5e), the simplified forms of equations (3a) – (3c) are written as follows:

$$\eta_1 = \left(\frac{G_f}{G_m}\right)(7 + 5\nu_f)(7 - 10\nu_m) \tag{6a}$$

$$\eta_2 = \left(\frac{G_f}{G_m}\right)(7 + 5\nu_f) \tag{6b}$$

$$\eta_3 = \left(\frac{G_f}{G_m}\right)(8 - 10\nu_m) \tag{6c}$$

Now using equations (4) and (5), equations (2a) – (2c) are simplified to give the following forms:

$$A = \left(\left(\frac{G_f}{G_m}\right)^2 (7 + 5\nu_f)\right) \cdot \begin{pmatrix} 8(4 - 5\nu_m)(7 - 10\nu_m)\phi_f^{\left(\frac{10}{3}\right)} - 50(7 - 12\nu_m + 8\nu_m^2)\phi_f^{\left(\frac{7}{3}\right)} + \\ 252\phi_f^{\left(\frac{5}{3}\right)} - 50(7 - 12\nu_m + 8\nu_m^2)\phi_f + 8(4 - 5\nu_m)(7 - 10\nu_m) \end{pmatrix} \tag{7a}$$

$$B = \left(\left(\frac{G_f}{G_m}\right)^2 (7 + 5\nu_f)\right) \cdot \begin{pmatrix} -4(1 - 5\nu_m)(7 - 10\nu_m)\phi_f^{\left(\frac{10}{3}\right)} + 100(7 - 12\nu_m + 8\nu_m^2)\phi_f^{\left(\frac{7}{3}\right)} \\ -504\phi_f^{\left(\frac{5}{3}\right)} + 150(3 - \nu_m)\nu_m\phi_f + 6(4 - 5\nu_m)(15\nu_m - 7) \end{pmatrix} \tag{7b}$$

$$C = \left(\left(\frac{G_f}{G_m}\right)^2 (7 + 5\nu_f)\right) \cdot \begin{pmatrix} 4(5\nu_m - 7)(7 - 10\nu_m)\phi_f^{\left(\frac{10}{3}\right)} - 50(7 - 12\nu_m + 8\nu_m^2)\phi_f^{\left(\frac{7}{3}\right)} \\ +252\phi_f^{\left(\frac{5}{3}\right)} + 25(\nu_m^2 - 7)\phi_f - (7 + 5\nu_m)(8 - 10\nu_m) \end{pmatrix} \tag{7c}$$

It should be noted that the term $[(G_f / G_m)^2 (7 + 5\nu_f)]$ is common to the simplified expressions for A, B, and C as given in the above equations (7a) – (7c). Hence, when these are substituted in equation (1), the dependence of (G_c / G_m) on G_f, G_m and ν_f ceases to exist. The only dependence that is left is on ν_m as shown below.

Proposed Simplified Model

On using the simplifications as discussed above, the proposed simplified model is written as:

$$A_1\left(\frac{G_c}{G_m}\right)^2 + B_1\left(\frac{G_c}{G_m}\right) + C_1 = 0 \tag{8}$$

where

$$A_1 = \begin{pmatrix} 8(4 - 5\nu_m)(7 - 10\nu_m)\phi_f^{\left(\frac{10}{3}\right)} - 50(7 - 12\nu_m + 8\nu_m^2)\phi_f^{\left(\frac{7}{3}\right)} + 252\phi_f^{\left(\frac{5}{3}\right)} \\ -50(7 - 12\nu_m + 8\nu_m^2)\phi_f + 8(4 - 5\nu_m)(7 - 10\nu_m) \end{pmatrix} \tag{9a}$$

$$B_1 = \begin{pmatrix} -4(1-5\nu_m)(7-10\nu_m)\phi_f^{\left(\frac{10}{3}\right)} + 100(7-12\nu_m+8\nu_m^2)\phi_f^{\left(\frac{7}{3}\right)} \\ -504\phi_f^{\left(\frac{5}{3}\right)} + 150(3-\nu_m)\nu_m\phi_f + 6(4-5\nu_m)(15\nu_m-7) \end{pmatrix} \tag{9b}$$

$$C_1 = \begin{pmatrix} 4(5\nu_m-7)(7-10\nu_m)\phi_f^{\left(\frac{10}{3}\right)} - 50(7-12\nu_m+8\nu_m^2)\phi_f^{\left(\frac{7}{3}\right)} \\ +252\phi_f^{\left(\frac{5}{3}\right)} + 25(\nu_m^2-7)\phi_f - (7+5\nu_m)(8-10\nu_m) \end{pmatrix} \tag{9c}$$

The stiffness ratio (G_c / G_m) can be obtained from the quadratic equation (8) and making a deliberate choice of the positive root, as

$$\left(\frac{G_c}{G_m}\right) = \left(\frac{\sqrt{(B_1^{\,2} - 4A_1C_1)} - B_1}{2A_1}\right) \tag{10}$$

Micromechanical models can predict fundamental material properties of a composite based upon the properties of the individual constituents with due consideration of their relative properties and geometric information of the various constituent phases. Equations (1) – (3) conform to a micromechanical model because the stiffness ratio (G_c / G_m) is dependent on all constituent properties G_f, G_m, ν_f and ν_m as well as on the volume fraction of the filler ϕ_f . The simplified form as given by equations (8) – (10), however, shows that the stiffness ratio (G_c / G_m) is dependent on only one constituent property ν_m and of course, on the volume fraction of the filler ϕ_f. This implies that the stiffness ratio (G_c / G_m) is basically insensitive to the other constituent properties, namely, G_f, G_m and ν_f. With this being the case, the predictions from equations (1) – (3) must match closely with those of equations (8) – (10), irrespective of the values of G_f, G_m and ν_f. This was checked for different reasonably realistic values of G_f, G_m and ν_f and found to be indeed true. The proposed simplified model gave almost identical predictions to those of the Christensen and Lo model [13] for chosen realistic values of the various parameters. This confirmed that the approximations used in simplifying the model were correct and did not lead to any significant errors.

The results from the model when compared with experimental data shown in Figure 2 indicate that the theoretical predictions fall short of the experimental data. This is on account of the fact that the GSCS model in the present form is unable to capture the effect of all the variables involved in the increase of the stiffness ratio of the mastics. The model is being applied over a range of volume fractions including the higher range at which the assumptions of Christensen and Lo [13] do not hold. This has been pointed out by Alberola and Mele [23] who extended the GSCS model to a wider range of volume fraction of fillers by considering the percolation threshold in the model using the percolation theory, which is discussed, in the subsequent section. For a volume fraction of fillers below the percolation threshold, the matrix is continuous. At the maximum volume fraction of the fillers, the fillers form the continuous phase. At intermediate volume fraction of the fillers, only some particles touch one another and form local continuous phases. The micromechanical model in the form given by equations (1) – (3) as well as the simplified form given by equations (8) – (10) do not account for these changes in the continuous phase with increasing volume fraction of the fillers. Thus, to obtain realistic results from the micromechanical model, it would be necessary to invoke the concepts of the percolation theory and present a modified form of the model.

Percolation Theory

Percolation theory basically deals with the connectivity of components in a system. The credit for the first time application of the concept of percolation theory goes to Hammersley [24]. Since then, there has been a continued interest in the use of this concept to a wide variety of composite systems. The equations of state for concentrated suspensions were obtained through this concept by Kang and Eringen [25] and applied to blood rheology. Interesting correlations between percolation threshold and elastic properties of block copolymers have been proposed by Hsu et al [26]. The percolation concept was used by Ouali et al [27] in the phenomenological law of Takayanagi et al [28] in order to account for an effective content of the viscoelastic medium in composites. The microstructure of cement-based materials also provides numerous examples of percolation phenomena [29]. Many such examples can be cited wherein the percolation theory is applied to various composite systems, including pavement materials and soils [30]. If one goes through the fundamentals of the percolation theory, it is quite obvious that the concept would as well work for asphalt mastics.

An excellent introduction to the percolation theory is provided by Stauffer and Aharony [31]. The essence of the percolation theory is to determine how a given set of sites, positioned in a regular or random manner in space, is interconnected according to a defined bonding criterion. At the percolation threshold, i.e. for a critical site fraction, a topological singularity occurs corresponding to a connection between sites on a large scale.

At low volume fractions of fillers, all particles are randomly distributed within the matrix, which forms a continuous phase. The matrix is then described as the percolated phase. With increasing volume fraction of filler ϕ_f, there is increasing probability of the particles touching one another and in this process part of the matrix gets entrapped within clusters of particles thereby progressively decreasing the volume fraction of the percolated matrix ϕ_{pm}. The schematic diagram of the entrapped non-percolated matrix in a cluster of particles embedded in a percolated matrix is shown in

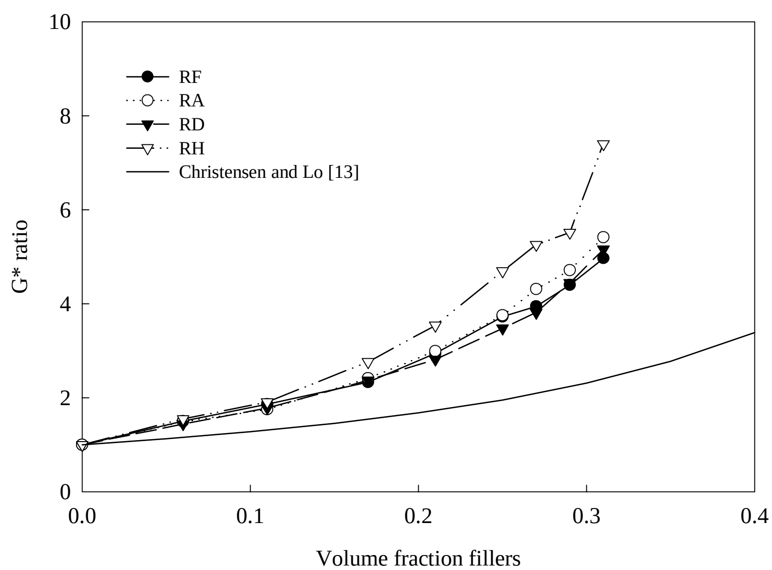


Figure 2. Comparison of the predictions of the Christensen and Lo model [13] with experimental results. The mastics were made with asphalt ABM-1 and aggregates from the materials reference library.

Figure 3. This process begins at a critical value of the volume fraction of the filler ϕ_{fpt}, which is termed as the percolation threshold. It is possible to determine the percolation threshold theoretically and this value has been found to vary from 0.15 [26, 32] to 0.30 [33, 34] in a 3-D lattice model. In reality, of course, this critical value can vary outside the theoretical bounds because of its dependence on various parameters such as, filler particle size, shape, size distribution, orientation, wetting behavior as well as matrix viscosity [34-36]. Hsu et al [26] suggest that this value ought to be determined empirically in order to be realistic. Alberola and Mele [2] chose a value of 0.2 because their experimental data showed a strong decrease in the damping factor at volume fractions of filler greater than 0.2.

There is an upper limit to the volume fraction of the fillers beyond which no more filler can be packed in a given volume. This is termed as the maximum volume fraction of the filler ϕ_{fmax}. Depending on the method of filler packing, the value of ϕ_{fmax} can vary from 0.52 for cubic structure of uniform hard spheres to 0.74 for a hexagonal structure of uniform hard spheres [37]. Choosing an appropriate size distribution can significantly increase the values of ϕ_{fmax}. At the maximum volume fraction of the filler, the particles are connected on a large scale. The filler is then percolated, while the matrix is nonpercolated because all of it is entrapped within the filler and remains unconnected. The volume fraction of the nonpercolated matrix ϕ_{mmin} ($= 1 - \phi_{fmax}$) is considered to be the minimum level for the matrix beyond which it would always be percolated in varying degrees. The matrix will be completely percolated at and below the percolated threshold volume fraction of the filler. Thus, the matrix is partially percolated between the following limits when $\phi_{mmin} < \phi_m < 1 - \phi_{fpt}$. The volume fraction of the percolated matrix is defined from the percolation theory [31] as

$$\left(\frac{\phi_{pm}}{\phi_m}\right) \approx (\phi_m - \phi_{mmin})^{0.4} \tag{11}$$

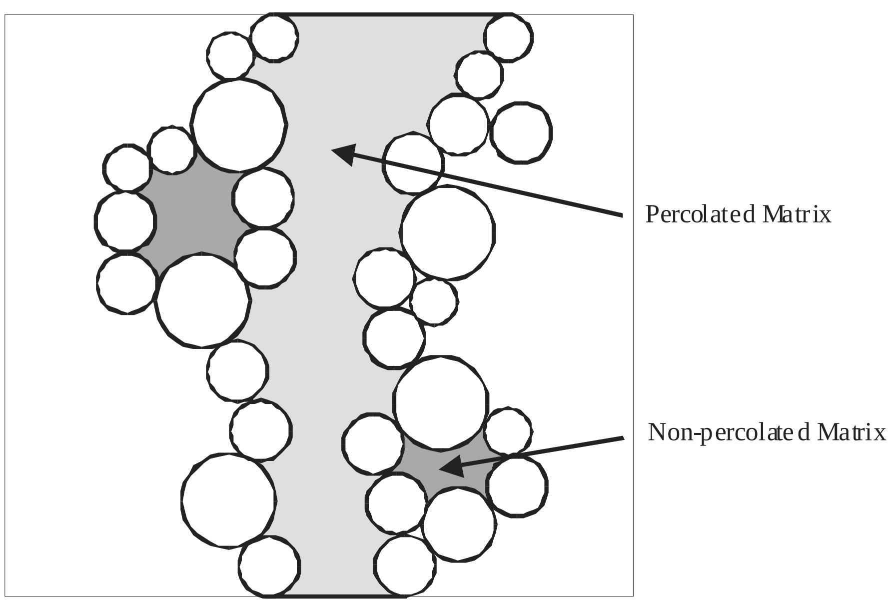


Figure 3. Schematic diagram of the entrapped non-percolated matrix in a cluster of particles embedded in a percolated matrix.

The boundary conditions imposed on the above equation (11) are the following:

$$\phi_{pm} = 0 \text{ for } \phi_m = \phi_{m\min} \text{ and } \phi_{pm} = \phi_m \text{ for } \phi_m = \left(1 - \phi_{fpt}\right) \tag{12}$$

Combining equations (11) and (12) gives

$$\phi_{pm} = \phi_m \left(\frac{\phi_m - \phi_{m\min}}{1 - \phi_{fpt} - \phi_{m\min}} \right)^{0.4} \tag{13}$$

Equation (13) is thus rewritten in terms of the effective volume fraction as

$$\phi_{eff} = 1 - (1 - \phi_f) \left(\frac{1 - \dfrac{\phi_f}{\phi_{f\max}}}{1 - \dfrac{\phi_{fpt}}{\phi_{f\max}}} \right)^{0.4} \tag{14}$$

It is now assumed that, because a certain amount of nonpercolated matrix is entrapped within the filler particles, the system behaves as though the volume fraction of the filler is greater than the actual value. This is termed as the effective volume fraction of the filler ϕ_{eff} and is calculated as 1- ϕ_{pm}. This is different from the actual volume fraction of the filler ϕ_f, which defined as 1- ϕ_m.

Proposed Modified Model

The proposed simplified model given by equations (8) – (10) is now modified by replacing ϕ_f by ϕ_{eff} to give the final form of the modified model as

$$\left(\frac{G_c}{G_m} \right) = \left(\frac{\sqrt{B_2^2 - 4A_2C_2} - B_2}{2A_2} \right) \tag{15}$$

where

$$A_2 = \left(\begin{array}{l} 8(4 - 5\nu_m)(7 - 10\nu_m)\phi_{eff}^{\left(\frac{10}{3}\right)} - 50(7 - 12\nu_m + 8\nu_m^2)\phi_{eff}^{\left(\frac{7}{3}\right)} \\ +252\phi_{eff}^{\left(\frac{5}{3}\right)} - 50(7 - 12\nu_m + 8\nu_m^2)\phi_{eff} + 8(4 - 5\nu_m)(7 - 10\nu_m) \end{array} \right) \tag{16a}$$

$$B_2 = \left(\begin{array}{l} -4(1 - 5\nu_m)(7 - 10\nu_m)\phi_{eff}^{\left(\frac{10}{3}\right)} + 100(7 - 12\nu_m + 8\nu_m^2)\phi_{eff}^{\left(\frac{7}{3}\right)} \\ -504\phi_{eff}^{\left(\frac{5}{3}\right)} + 150(3 - \nu_m)\nu_m\phi_{eff} + 6(4 - 5\nu_m)(15\nu_m - 7) \end{array} \right) \tag{16b}$$

$$C_2 = \left(\begin{array}{l} 4(5\nu_m - 7)(7 - 10\nu_m)\phi_{eff}^{\left(\frac{10}{3}\right)} - 50(7 - 12\nu_m + 8\nu_m^2)\phi_{eff}^{\left(\frac{7}{3}\right)} \\ +252\phi_{eff}^{\left(\frac{5}{3}\right)} + 25(\nu_m^2 - 7)\phi_{eff} - (7 + 5\nu_m)(8 - 10\nu_m) \end{array} \right) \tag{16c}$$

and ϕ_{eff} is given by equation (14). In the proposed simplified model given by equation (10), (G_c/ G_m) is a function of only two parameters ν_m and ϕ_f. It can be seen that in the proposed modified model, (G_c / G_m) is a function of the parameters ν_m, ϕ_f, ϕ_{fpt} and ϕ_{fmax}. The additional parameters ϕ_{fpt}

and ϕ_{fmax} provide the much needed flexibility to match the theoretical predictions with experimental data. Use of adjustable parameters in empirical equations is quite common in the literature on the rheology of filled systems [38]. However, in the present case, the equation itself is derived through micromechanical modeling unlike an empirical equation, and the adjustable parameters, too, have a basis for their presence because of their derivation from the percolation theory.

The proposed modified model has better validity at higher volume fractions on account of the use of the percolation theory. At low volume fractions where the equation may not hold well, dilute suspension results of equations (1) – (3) may be invoked as given by Christensen and Lo [13]

$$\left(\frac{G_c}{G_m}\right) = 1 - \left(\frac{15(1-\nu_m)(1-\frac{G_f}{G_m})\phi_f}{(7-5\nu_m) + 2(4-5\nu_m)\frac{G_f}{G_m}}\right) + O(\phi_f^{\,2}) \quad \text{for} \quad \phi_f \to 0 \tag{17}$$

Again using the approximation given in equation (5) when $G_f >> G_m$, the simplified form of equation (17) is written as

$$\left(\frac{G_c}{G_m}\right) = 1 + \left(\frac{15(1-\nu_m)\phi_f}{2(4-5\nu_m)}\right) \quad \text{for} \quad \phi_f \to 0 \tag{18}$$

The above equation can be used at low volume fractions (ϕ_f<0.1), where equation (15) may not provide good predictions.

Experimental

Asphalt and aggregates from the materials reference library (MRL) were used in this study. The aggregates were ground, and the material passing –200 mesh was used as fillers in these experiments. The specific gravity of the ground fillers was measured according to AASHTO T133 using kerosene. The asphalt was weighed in a 4-oz can, and the appropriate amount of filler was added to it to obtain the required volume fraction of fillers. The can was heated at 163°C for 15 minutes and stirred with a mechanical stirrer for one minute. The mastic was poured into a test tube for settling experiments in order to estimate actual ϕ_{fmax} and two pellets poured into silicone rubber molds for the dynamic shear rheometer (DSR) measurements similar to what was done in an earlier paper [39]. The DSR measurements were typically completed within two hours of pouring the pellets. Mastics with filler contents of 0, 6, 11, 17, 21, 25, 27, 29, 31 percents (by volume) were prepared. Care was taken to ensure that all the mastics and the blank were made using asphalt from the same 1-quart can. This was crucial since different thermal histories in different cans would cause slight differences in rheological properties that would thwart further analyses.

Strain sweeps were run at 25°C and 58°C using 8-mm and 25-mm parallel plates, respectively, using a Rheometrics RDA II dynamic shear rheometer. The strain levels were chosen such that a minimum torque of 0.2 gm-cm and a maximum of 200 gm-cm were obtained. Complex shear modulus and phase angle at a strain in which all the mastics were within the linear viscoelastic range were chosen for further analysis.

Results and Discussion

The numerical results predicted by various models were carried out in mathematical software (Mathcad Professional 8). Figure 4 shows the prediction of our proposed simplification (Equations 8-10) of the model of Christensen and Lo [13]. Comparison of the predictions of our proposed simplified model with the original from Christensen and Lo [13] shows that the error due the simplification is very low for low matrix stiffness. When the shear modulus of the binder is very high, say 100 MPa (on the order of stiffness measured with a bending beam rheometer), the stiffening ratio predicted by Christensen and Lo model [13] is slightly smaller. Dependence of the stiffening ratio on the modulus of the matrix is not present in the proposed simplified model. Even at high matrix modulus the simplification causes not more than 4% error at the highest volume fractions. As we shall see further, at these high filler concentrations, the simplified and original Christensen and Lo model [13] may not be applicable.

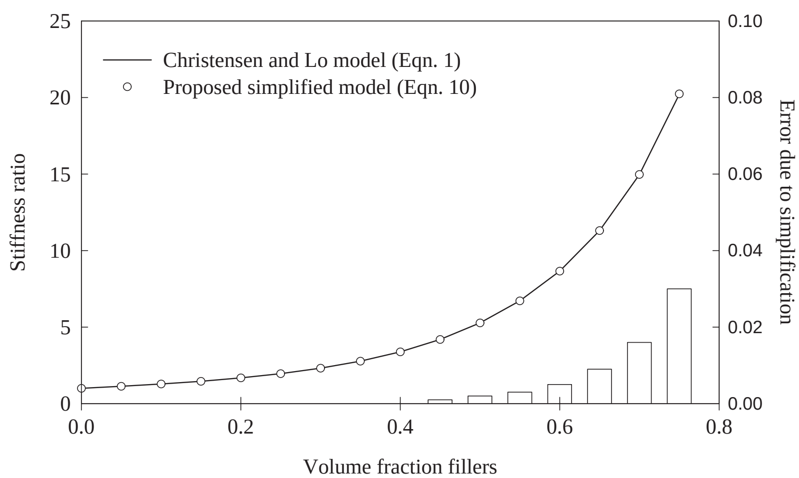


Figure 4. Comparison of the theoretical predictions of the Christensen and Lo model [13] with that of the proposed simplified model.

In Figure 2, it was shown that the Christensen and Lo model [13] underestimates the stiffness of the composites for typical asphalt-filler systems. The predictions fall short because the model fails at higher volume fraction of the filler. Along the lines of Alberola and Mele [23], the percolation theory was applied to the simplified Christensen and Lo model to yield a modified model, which is much simpler in form than that used by Alberola and Mele [23]. The numerical results predicted by the proposed modified model (Equation 15) are plotted in Figure 5. These results are consistent with those obtained by Alberola and Mele [23] showing that the proposed modified model, which is available in a much simpler form, can be used instead.

There is one major difference in the Alberola and Mele model [23] and the proposed modified model. In the proposed modified model, there is no dependence on particle size. However, in the Alberola and Mele model [23], there is a term where the radius of the second phase is input and drives the equation to predict higher stiffening at higher particle sizes. This is not consistent with data observed in the literature [38]. Typically, smaller the particle size, the stiffer the composite due to several effects, which disappear when a suitable surfactant is added [41]. Thus, this model by Alberola and Mele [23] shows a particle size dependence that is contrary to that observed by researchers. Christensen and Lo model [13] does not depend on the particle size of the fillers. Since

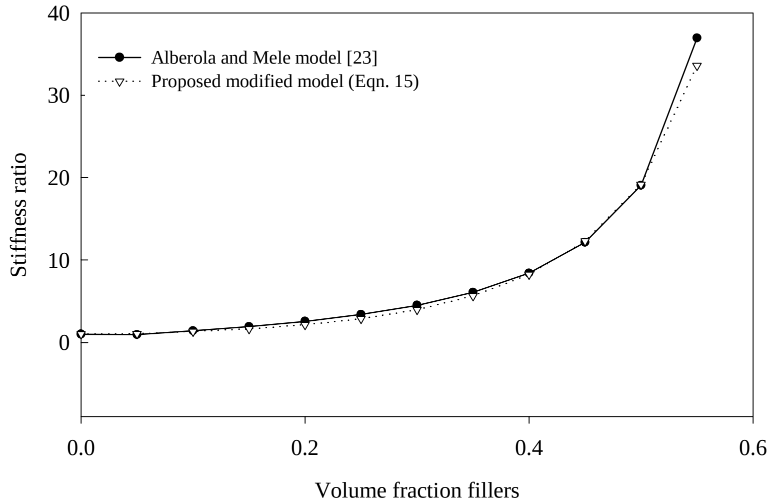


Figure 5. Comparison of the predictions of the proposed modified model with those of Alberola and Mele [23].

our derivation has its basis on the Christensen and Lo model [13], our proposed models also do not have a particle size dependence.

The dependence of the stiffening on the parameters, ϕ_{fpt} and ϕ_{fmax}, are shown in Figure 6. In Figure 6A, ϕ_{fpt} was kept constant while ϕ_{fmax} was changed from 0.45 to 0.70. These curves show the same dependence of modulus on ϕ_{fmax} in the proposed simplified model that was seen in Nielsen's equation [40]. The ϕ_{fmax} is the vertical asymptote for these curves. At ϕ_{fmax} the modulus of the composite approaches infinity. Figure 6B shows the modulus as a function of ϕ_{fpt}. This parameter does not have as much effect on the composite modulus as ϕ_{fmax}. This parameter affects the instantaneous slope of the curve. The higher the ϕ_{fpt}, the lower is the slope. This behavior is inverse of the parameter K_E in the Nielsen's equation [40].

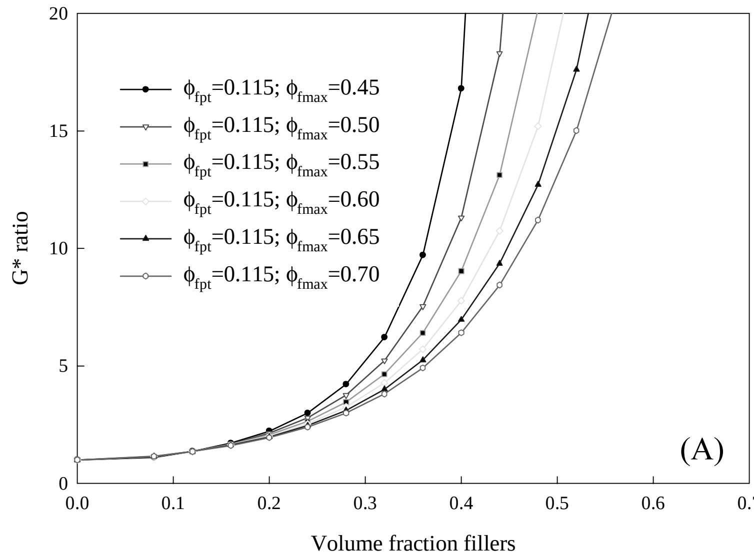
The success of any model depends on how well the model fits the experimental results and whether it is consistent with the laws/theories of physics. Equation 15 has two adjustable parameters— ϕ_{fpt} and ϕ_{fmax}. By choosing the appropriate values for these parameters the model can be made to fit the data well. The results are illustrated in Figure 7. The parameters for the four asphalt-filler systems studied are listed in Table 1. For comparison, the ϕ_{fmax} estimated by fitting Nielsen's equation to the data is also given. ϕ_{fmax} estimated from Nielsen's equation was found to agree with ϕ_{fmax} measured by settling experiments [39] and can be taken to represent the true value.

The data was fitted with Equation 15 using a trial and error procedure. The values for ϕ_{fmax} predicted by Equation 15 and those that were obtained from Nielsen's equation agree with one another for fillers RD and RH but do not agree for RF and RA. Also, the values of ϕ_{fpt} do not seem to be realistic. One would expect values of ϕ_{fpt} should be around 0.10 and 0.15. It is not realistic that a composite would become percolating by addition of just 2% fillers by volume. It was found that the model was quite sensitive to the value of ϕ_{fpt}; hence setting re-

Table 1. Aggregates used in this study

Aggregate	Code	ϕ_{fpt}	ϕ_{fmax}	ϕ_{fmax}*
Glacial gravel	RF	0.10	0.55	0.44
Granite	RA	0.05	0.60	0.50
Limestone	RD	0.03	0.45	0.46
Graywacke	RH	0.02	0.48	0.44

* Predicted by Nielsen's equation



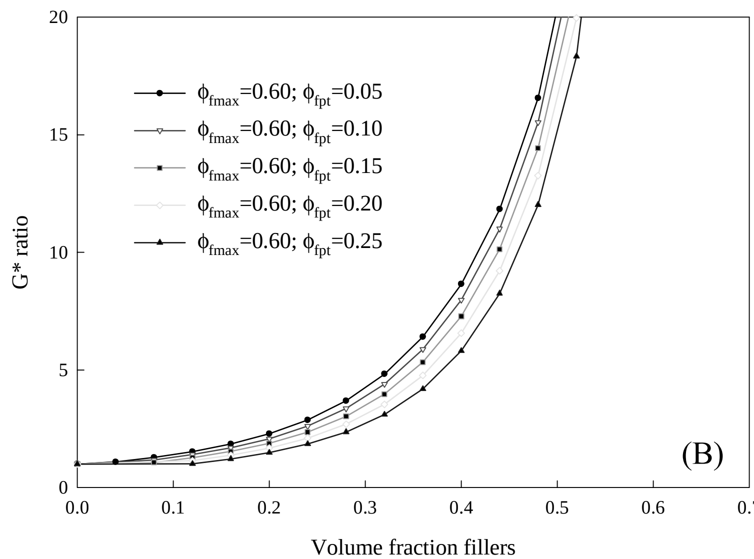


Figure 6. The dependence of the stiffening on (A) the percolation threshold ϕ_{fpt} and (B) the maximum packing fraction ϕ_{fmax}.

alistic values offsets the fit of the model to the measured data.

Unfortunately, the percolation limit cannot be measured experimentally unless the filler-matrix system had special properties. Alberola and Mele [23] point out that these can best be estimated empirically and just use a value of 0.20.

Application to Asphalt Mastics

The models described herein are very useful in developing an understanding of how the various properties of the fillers and the asphalt matrix affect the stiffening of two-phase asphalt mastics. Such an understanding will help in engineering the properties of asphalt-filler systems according to requirements. However, applying these models to fit experimental data is not simple, because the models themselves are too complicated despite the simplifications suggested. This is the reason

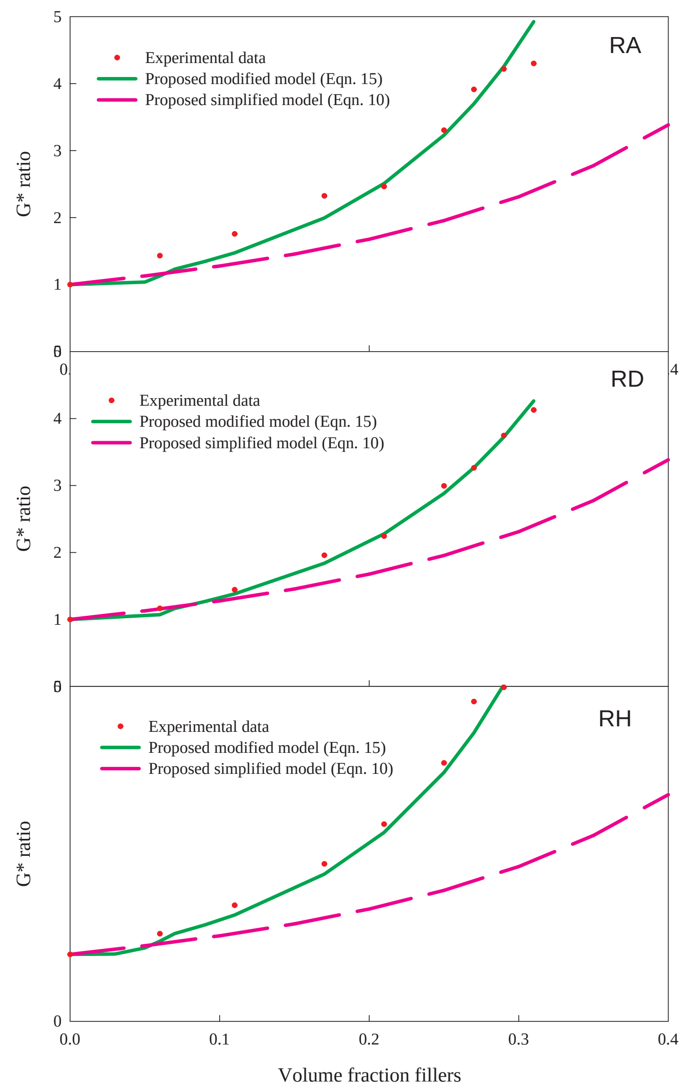


Figure 7. The fit of the proposed models to experimental data.

why several empirical equations, such as the Maron-Pierce equation and others, are in use for composite systems [38]. Some such equations even yield parameters that reflect physical properties of the system. For example, the Nielsen's equation relates the changes in stiffness due to the addition of fillers to parameters such as maximum packing fraction, ϕ_{fmax}, and generalized Einstein coefficient, K_E, which have been verified to be real physical parameters through independent technique [40].

The concept of the use of percolation theory to micromechanical models to make the models fit the experimental data is perhaps appropriate. This is because the various factors that affect the stiffening of the matrix by the fillers, such as particle shape, average particle size, particle size distribution, degree of agglomeration, etc., would also intuitively affect the percolation limit. In this way, these factors are accounted for in micro-mechanical models.

However, the micro-mechanical model tends to be complicated even though efforts were taken to simplify the model. Even if it were possible to fit the data to the proposed model, it is difficult to understand a particular data set by using this equation alone. This is due to the following reasons:

1. The uncertainty in the estimation of the parameters, ϕ_{fpt} and ϕ_{fmax}, is likely to be high due to the complex nature of the equation. In such cases, it may not be possible to compare the behavior of one asphalt-filler system with another with a great deal of certainty.
2. At the present time, the relationship between the ϕ_{fpt} and particle shape, size, size distribution, degree of agglomeration etc. are not clearly known. These relationships are important if we are to use the model to design mastics with particular property requirements.

Micromechanical models discussed here require the experimental determination of quantities such as ϕ_{fpt} and ϕ_{fmax}. The methods to determine these quantities to required precision are not simple. Such methods should be developed before we can take advantage of these methods.

Conclusions

The following conclusions can be drawn from this study:

1. The Christensen and Lo model [13] can be simplified to a form applicable to asphalt mastics. The simplified model predicts stiffening ratios that are independent of matrix stiffness.
2. The percolation theory could be effectively incorporated into the proposed simplified model [Equation (10)]. The proposed modified model [Equation (15)] gives predictions similar to Alberola and Mele [23], but the proposed equation is much simpler in form.
3. Alberola and Mele model [23] shows a dependence of stiffness ratio on particle size that is contrary to experimental data. The proposed modified model [Equation (15)] gives a stiffness ratio that is independent of particle size.
4. Although micro-mechanical models are important in understanding the moduli of composite systems, they are cumbersome for use from a practical viewpoint. Semi-empirical models such as Nielsen's model would probably serve better.

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