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Micromechanical shear modulus modeling of activated crumb rubber modified asphalt cements

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HIGHLIGHTS

- Absorption of the lighter fractions by the crumb rubber particles.
- A method to quantify the swelling and splitting of the rubber particles.
- Use of micromechanical models to accurately predict the dynamic shear modulus of rubberized asphalt cement.

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ABSTRACT

The rheological properties of three asphalt cements containing reacted and activated rubber (RAR) are evaluated to quantify the relative effects of swelling, splitting, and absorption. Rheological testing and electron microscopy are used to measure the dynamic shear modulus, $|G^*|$, and rubber particle changes respectively. It is found that $|G^*|$ increased for all RAR and that particles swell 15.8–49.3% with those in softer asphalt showing the greatest swelling. Micromechanical models are used to predict $|G^*|$ of the materials. The Hashin and Christensen models are found to accurately predict the measured moduli after accounting for the swelling, splitting, and absorption.

1. Introduction

The use of crumb rubber with and without physical and chemical modification to improve the engineering performance of asphalt concrete has been well documented over the last 40 years [1–8]. Within these documented studies, the performance benefits from incorporating rubber are: greater rutting resistance, mitigation of thermal reflective cracking, increased resistance to fatigue cracking, and noise reduction. It is often cited that the reason for the increased performance of these rubber modified asphalts is that during the blending process, the crumb rubber particles absorb the aromatic oils and resins from the asphalt cement leaving the binder with a higher concentration of asphaltenes while at the same time storing the lighter resins for times when they are needed for example to accelerate healing. This diffusion of lighter fractions is postulated to result in swelling of the rubber particles in a time and temperature dependent process that can have a substantial impact on the volume occupation and thus performance [9,10]. The concept of rubber swelling is somewhat controversial. It is documented that crumb rubber particles can swell by 3–5 times their original size [9,11,12], but very little peer reviewed literature has directly quantified the precise volume increase. Kutay et al. quantified the structural change in crumb rubber modified asphalt by using X-ray microtomography and image analysis and found that the volumetric percentage of rubber was less after the mixing process than was originally added. However, the researchers also suggested that this volume decrease was an artifact of melting or splitting of the crumb rubber during mixing and thus concluded that the crumb rubber particles will increase their size two to three times of their original size [13]. Airey et al. also studied this phenomenon using a basket drainage method to first separate the binder and rubber particles after mixing with rubber and

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performed rheological testing as well as asphaltene content test (did not specified which method they used) on the pre-blending and post–blended asphalt to measure the loss of SARA fractions during mixing. Based on this testing, they concluded that the change in mass of crumb rubber particles was independent of the original source of the asphalt and only marginally dependent on the penetration grade of the asphalt cement used. Specifically, they found that the post-rubber mixing residue from the low penetration asphalt had a greater amount of lighter fractions remaining and thus the rubber in these asphalts must have taken in less of these fractions [9]. Both studies provide evidence of swelling and degradation of the rubber during mixing, but have some limitations. In the Kutay study, the authors must infer that swelling is negated by splitting without directly quantifying the number of particles that have split or disintegrated. The Airey et al. investigation did not directly measure the swelling and inferred its extent based on only mass changes.

The importance of estimating the swelling of rubber particles in asphalt is that if known, then it may be possible to apply established computational and analytical composite models with some level of accuracy to estimate the modulus of the crumb rubber modified asphalt cement and by consequence design rubber blends for maximum benefit. Some models to describe the effect of rubber particles in asphalt (notably as a function of diameter and surface area) have been developed [14]. However, these approaches are based on empirical correlations between rubber related variables and resultant composite response, which limit their ability to provide direct and generalized insight into the impact of swelling and other physical phenomenon. Conversely, there are many micromechancial models, which do have the potential to provide such insights, that have been used to predict the viscosity and/or shear modulus of filled, viscous composites like rubber modified asphalt [15]. The models themselves attempt to reflect the impacts of the volume related impacts of particulates in a binding matrix using analogous physical systems. This, interpretation of the predictions in systems where particle actively modify the binding matrix can be challenging. The most well-known of these models is probably the Einstein function. This function models the system assuming non-interacting rigid spherical particles that are highly dispersed in a fluid medium. Physically this model is representative of systems that have very low particulate concentrations (<5%), and studies suggest that the intrinsic viscosity from the function is not constant and it is related to the physical characteristics of the mineral filler ranging from 2.5 to 4.9 [16]. Others have offered additional modifications to this function, for example Roscoe modified this model by considering the different sizes of mineral fillers [17]. Despite this and other modifications studies agree that the composite models derived from Einstein's original function work well for some suspension or matrix but do not show an accurate representation of asphalt systems and in most cases under predict the modulus of the composite [16,18–21]. However, such under predictions can be the construed because of the model missing important physical parameters. Bahia et al. [12] used the Einstein and Mooney model to predict the viscosity of crumb rubber modified asphalt cement. As other studies have suggested, the models underestimated the viscosity values, and the authors used this finding as an indication that the interaction between the rubber and asphalt is not one involving simple solid inclusions. Consequently Bahia et al. propose that there must be a swelling mechanism and a change in the matrix (i.e. asphalt cement) [19]. Jamrah et al. used the Rule of Mixtures, Inverse Rule of Mixtures and the Differential model to backcalculate the modulus of the swollen rubber assuming that each model correctly described the composite system. Citing the previous study from Kutay et al. the volume concentration of rubber was adjusted to 300% of the original volume [22]. However, these models (as well as the Einstein derived forms) have been shown to systematically underestimate the stiffening behavior of a related system consisting of asphalt cement and mineral particles smaller than 0.075 mm (known as asphalt mastic) [20,21,23]. In this system, the particulate phase does not swell or change its properties in the presence of the asphalt and yet, the models show a systematic under prediction of the composite modulus.

Thus, while most studies agree about the primary mechanisms of interaction between the asphalt cement and the crumb rubber, that is the preferential absorption and the swelling. They differ in quantifying the volume expansion and linking these mechanisms to the modulus of the composite system. Consequently, it is difficult to know what course of action to make when choosing or designing a rubber blend in order to maximize the potential performance benefit. The objective of this paper strives to fill in this knowledge gap in understanding the multi-physical phenomena in asphalt mastics by reporting on a research study with the following objectives; 1) to evaluate the rheological properties of three asphalt cements modified with reacted and activated rubber (RAR), 2) estimate rubber swelling and potentially particle splitting, and 3) to quantify the relative effects of particle swelling, absorptions, and swelling/splitting in a composite system consisting of asphalt cement and RAR.

2. Micromechanical models

Although many functions and techniques exist to calculate and predict the response of asphalt composites, analytical micromechanical models that are based on the material properties (Poisson's ratio, modulus and volume fraction) and particle interaction via effective medium methods have proven to be most successful [20]. Many such models exist, but the most well-known and the ones used in this study are the Hashin Composite Sphere model, Eq. (1), and the Christensen model, Eq. (2). Both of these models are classified as effective medium models because they essentially solve the mathematical problem of a single particle within an effective medium, which itself has properties that are consistent (either in average energy or average displacement) with the matrix of the real composite. Note that the Christensen model has the form of a quadratic function, and that the parameters *A*, *B*, and *C* are calculated from known values [24]. Once these parameters are calculated, then the quadratic formula is used to calculate the composite shear modulus. It should be noted that other models were also evaluated during the study including the Einstein model [25], the Roscoe model [26], Mori-Tanaka scheme developed as applied by Benveniste [27], and the Differential method [28,29]. These models yielded values very similar to the Hashin and Christensen result and are not shown here in the interest of brevity, but their formulations are shown in the Appendix.

$$\frac{|G^*|_c}{|G^*|_b} = 1 + \frac{15(1-\nu_m)\left(\frac{G_r}{|G^*|_b} - 1\right)C_\nu}{7 - 5\nu_m + 2(4-5\nu_m)\left[\frac{G_r}{|G^*|_b} - \left(\frac{G_r}{|G^*|_b} - 1\right)C_\nu\right]} \tag{1}$$

$$A\left(\frac{|G^*|_c}{|G^*|_b}\right)^2 + B\left(\frac{|G^*|_c}{|G^*|_b}\right) + C = 0 \tag{2}$$

where G_c is the modulus of the composite in kPa, G_r is the modulus of the rubber [30] in kPa, G_b is the modulus of the matrix (the asphalt) in kPa, ν_r is the Poisson's ratio of the rubber (assumed equal to 0.5 for this study) [31,32], ν_m is the Poisson's ratio of the matrix, C_v is the volumetric concentration of the filler, and *A*, *B*, *C* are functions of G_r, G_b, ν_r, ν_m, and C_v (shown in Appendix).

The need and significance of using these models in this paper is that they permit a more direct interpretation of the multiphysical phenomenon that take place in the rubber based asphalt systems.