Evaluation of Moisture Sorption and Diffusion Characteristics of Asphalt Mastics Using Manual and Automated Gravimetric Sorption Techniques

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Abstract: One of the most important factors influencing the durability of asphalt mixtures is moisture-induced damage resulting from the presence and the transport of moisture in pavements. Moisture-induced damage is an extremely complicated phenomenon that is not completely understood but believed to be governed by the interaction of moisture with asphalt mix components (mastic and aggregates). The objective of this study was, therefore, to characterize the sorption and diffusion characteristics of asphalt mastic using gravimetric vapor sorption techniques. Moisture transport, in the hygroscopic region, in asphalt mastics was studied using both static and dynamic gravimetric vapor sorption techniques to determine equilibrium moisture uptake and diffusion coefficients as a function of aggregate and filler types. For the 25-mm diameter thin asphalt mastic films and the testing conditions (23°C and 85% relative humidity) considered, the kinetics of moisture uptake obtained were characteristic of Fickian diffusion with a concentration-dependent diffusion coefficient. Equilibrium moisture uptake and diffusion coefficient estimated from the static measurements were comparable and of the same order of magnitude as those from dynamic sorption techniques. Both measurement techniques ranked the mixes similarly which suggest either method could be used to characterize moisture transport in asphalt mastics. Equilibrium moisture uptake was relatively higher in mixtures containing granite aggregates compared with limestone aggregate. In contrast, the diffusion coefficient of limestone aggregate mastics was higher than granite. Thus an inversely proportional relationship exists between moisture uptake and diffusivity of the asphalt mastics studied. The results suggest moisture transport is a function of aggregate type and that both equilibrium moisture uptake and diffusion coefficient are useful in studying moisture susceptibility in asphalt mixtures. The effect of mineral filler type on diffusion coefficient was minimal in the mastics containing granite aggregate but relatively high in mastic samples containing limestone aggregates. Diffusion coefficient was found to increase with sample thickness which was unexpected because diffusion coefficient (in an isotropic material) is considered an intrinsic property that is independent of sample size. The results suggested anisotropic diffusivity can occur in asphalt mastics and could be attributed to factors including mineralogy, microstructure, air voids, and the tendency of the aggregates to settle at the bottom of asphalt mastic with time. In addition to characterizing moisture transport in asphalt mastics, the results presented in this paper will be useful as inputs for numerical simulation of moisture damage in asphalt mixtures.

CE Database subject headings: Asphalts, Asphalt pavements, Material Properties, Water, Diffusion
Author keywords: Moisture diffusion, Diffusion Coefficient, Asphalt Mastics, dynamic vapor sorption, relative humidity, limestone, granite, Fick's law

Introduction

The transport of moisture into and/or through asphalt mastic is of great interest because it has relevance to the physico-chemical characterization, numerical modeling, and fundamental understanding of the moisture-induced damage phenomenon in asphalt mixtures, which is important for designing durable bituminous pavements. A key parameter that characterizes moisture transport in a material is the moisture diffusion coefficient. The effects of the presence and transport of moisture within asphalt mixtures is a leading cause of moisture damage that is a major cause of pavement distress. This is because the loss of cohesion within and / or the loss of adhesion between asphalt mastic and aggregate are commonly regarded as the principal causes of moisture-induced damage (Terrell 1994, Airey and Choi, 2006). The effects of moisture diffusion into the asphalt mastic as well as the effects of moisture on the adhesive bond between asphalt and aggregate are directly related to moisture diffusion characteristics of the mixture. Moisture can reach the asphalt-aggregate interface and cause stripping by diffusing through the asphalt or mastic. In addition, the moisture in the mastic can profoundly affect the rheology and engineering properties of the mastic (Cheng et al 2003). The mechanism of moisture-induced damage is currently not completely understood and therefore, empirical methods are currently the only commonly available means of studying the phenomenon. The study of moisture diffusion, with its focus on molecular movement of water at the mastic aggregate interface, offers a more fundamental approach for better understanding of the moisture-induced damage problem than existing empirical characterization test methods.

Traditionally, the susceptibility of an asphalt mixture to moisture-induced damage has been evaluated using an index-based parameter such as stiffness or strength, obtained before and after specified simulated moisture-induced tests (Airey and Choi, 2002). While these traditional approaches have been successfully used to evaluate moisture susceptibility, these tests are empirical in nature, and therefore require experience before their results can be properly interpreted. Also, prediction based on such empirical tests could be contradictory for certain mixtures (Apeagyei et al. 2006). Furthermore, the methods do not address moisture-damage at a level that could enable a fundamental understanding of the problem because moisture transport characteristics of the mastic and aggregate components of asphalt mixtures have not been routinely evaluated. The lack of moisture transport data for asphalt mix components (mastics and aggregates) is a major hindrance to a fuller understanding of moisture damage in asphalt mixtures. The

problem of the limited number of studies focused on moisture transport in asphalt mastic is further compounded by the rather large variations in reported diffusion coefficient values as a result of the different testing conditions (temperature and relative humidity) and experimental approaches (gravimetric and psychrometric) used in their determinations (Table 1).

It can be seen from Table 1 that one technique that has been reported for evaluating moisture diffusion in asphalt mastic is the measurement of weight gain in specimens submerged under water at room temperature (Kassem et al. 2006 and Vasconcelos et al. 2006). The reported diffusion coefficient for these studies ranged from 10 to 24 x 10^{-12} m^2/s at 25°C.

Another technique used to measure diffusion coefficient in some previous studies (Kringos et al. 2008 and Arambula et al 2010) is a gravimetric method similar to what Vasconcelos and co-workers used but with specimens exposed to moisture vapor (15% and 85% RH). The reported moisture diffusion coefficient values ranging from 0.13 to 0.36 x 10^{-12} and 254.0 x10^{-12} m^2/s appear to differ significantly. It should be noted that while Kringos and co-workers assumed Fick's second law (unsteady state diffusion) to estimate diffusion coefficient, the set-up for Arambula and co-workers appears to satisfy Fick's first law (steady-state diffusion and hence moisture permeability coefficient). As previously mentioned and further discussed next, the differences in experimental set-up used in the past may have contributed to the differences seen in the reported diffusion coefficient values.

In general, moisture can be transported in a porous material in three different ways: 1) diffusion, 2) capillary flow, and 3) hydraulic flow, depending on moisture content and the driving potential. When moisture content of a material is in the hygroscopic region (0-95% RH) diffusion is the main mechanism of moisture flow with vapor pressure as the driving potential. Capillary flow is the dominant mode of moisture transport for moisture content in the super-hygroscopic region (95<RH<100) and capillary pressure in the pores of the material is the driving potential. For materials with moisture content in the super-saturated region, hydraulic flow is governed by Darcy's law with external pressure gradient (air pressure, water pressure, or gravitation) as the driving potential. In the super-saturated region all capillaries have been filled and therefore, no capillary pressure exists. Thus, it can be argued that the differences in reported asphalt mastic diffusion coefficient are due to the differences in testing conditions (diffusion and capillary flow) used by different investigators.

This study looks at moisture diffusion in asphalt mastic using gravimetric sorption techniques assuming classical Case I or Fickian diffusion and contributes to the understanding of moisture damage by providing equilibrium moisture uptake and Fickian diffusion coefficients that are useful for micro-mechanical characterization as well as numerical simulation of moisture-induced damage in asphalt mixtures. The moisture transport properties obtained in this study will also be useful as inputs for numerical models that simulate moisture-induced damage in asphalt mixtures.

In the current work, water vapor transport in asphalt mastics was studied using both static and dynamic gravimetric vapor sorption techniques to determine equilibrium moisture uptake and diffusion coefficient as a function of two aggregate and filler types. Asphalt mastic and aggregate form the two main components of asphalt mixtures with the mastic acting as the adhesive that binds the aggregates together. It was expected that the results from testing the asphalt mastic, may be of use in better understanding of moisture movement and associated moisture-induced damage in asphalt pavements by identifying factors influencing moisture diffusion in asphalt mastics.

Theory

Moisture Uptake Profiles

A moisture uptake profile describes the relationship between the amount of moisture (M_t) a hygroscopic material exchanges (absorbs or desorbs), at a given relative humidity and temperature, with time. If w_0 is the initial (dry) mass of a given material and w_t is the mass after time t, then the moisture uptake can be computed as the ratio of the amount of moisture absorbed at a given time to the initial dry mass of the sample at the beginning of the test (Eq. 1).

$$Mass\ uptake\ (\%) = M_t = \frac{w_t - w_0}{w_0} * 100 \quad (1)$$

For a material at given temperature and relative humidity, moisture uptake increases until it reaches a thermodynamic equilibrium at which point no further changes in moisture uptake occurs. The moisture content at thermodynamic equilibrium (M_∞) is called equilibrium moisture uptake. The time it takes a material to reach M_∞ as well as the ratio $\frac{M_t}{M_\infty}$ provide important insights into moisture transport (diffusion rate).

Fick's first law

Considering moisture content as the potential for mass transfer through a unit area of a section of an isotropic material, the mass flow of moisture per unit area or flux (F) is given by Fick's first law (Eq. 2).

$$F = -D\frac{\partial C}{\partial x} \quad (2)$$

where F is the mass flow of moisture per unit area (kg/s × 1/m^2), D is the effective diffusion coefficient (m^2/s), C is the water concentration (kg/m^3), and x is the distance (m) in the flow direction. If the concentration C of the system is changing, then Fick's second law is the applicable model estimating diffusion coefficient under a given experimental condition as discussed next.

Determination of diffusion coefficient

When a thin planar sample is maintained at a constant relative humidity and moisture sorption is occurring via the two planes (x=0 and x=l) of the sample and utilizing the kinetic sorption data, moisture diffusion coefficient can be determined based on the solution of Fick's second law (Eq. 3) for an infinite plate geometry under one-dimensional isothermal conditions assuming a constant diffusion coefficient, negligible swelling, initial conditions (Eq. 4) and boundary conditions (Eq. 5). The assumption of a thin plate means the majority of diffusion occurs in-plane and radial diffusion is negligible.

$$\frac{\partial C}{\partial t} = D\frac{\partial^2 C}{\partial x^2} \quad (3)$$

$$t = 0, C = C_0 \; for \; 0 < x < l \quad (4)$$

$$t > 0, C = C_1 \; for \; x = l \text{ and } \frac{\partial C}{\partial x} = 0 \; for \; x = 0 \quad (5)$$

where t is time, l is the sample thickness, C is the water concentration, C_0 is the initial uniform water concentration, and C_1 is the constant moisture concentration at the surface.

The solution to Eq. 3 under the aforementioned conditions is given by Eq. 6 (Crank, 1975) which can be solved numerically using the sorption data to obtain the diffusion coefficient of the mastics. This is

one of the most accurate models for predicting moisture diffusion coefficient and will be called the full-form equation in this paper.

$$\frac{M_t}{M_\infty} = 1 - \sum_{n=0}^{\infty} \frac{8}{(2n+1)^2\pi^2} e^{\frac{-D(2n+1)^2\pi^2 t}{l^2}} \quad (6)$$

where M_t is the amount of moisture absorbed after time t, n is an integer, and M_∞ is the maximum amount of absorbed moisture at the thermodynamic equilibrium under a given relative humidity and temperature, D is the diffusion coefficient, and l is plate thickness.

Simplified solutions to Fick's second law

Assuming Fickian diffusion, Eq. 6 can be simplified (Crank, 1975) as shown in Eq. 7.

$$\frac{M_t}{M_\infty} = \frac{4}{l}\sqrt{\frac{Dt}{\pi}} \quad (7)$$

Therefore, plotting $\frac{M_t}{M_\infty}$ against $\sqrt{t}$, for a moisture content ratio of at least 0.5 yields a straight line which could be used to estimate moisture diffusion coefficient using Eq. 8.

$$D = \frac{S^2}{16}\pi l^2 \quad (8)$$

where S is the slope of the plot of $\frac{M_t}{M_\infty}$ against $\sqrt{t}$, as described above.

Another simplified method, the half-time method, was employed to estimate diffusion coefficient of asphalt mastics. From Eq. 6, if the time $t_{0.5}$ is determined such that $\frac{M_t}{M_\infty} = 0.5$, then the value of $\frac{t}{l^2}$ for which $\frac{M_t}{M_\infty} = 0.5$ is given by Eq. 9 from which the diffusion coefficient (D) could be computed (Eq. 10).

$$\frac{t_{0.5}}{l^2} = -\left(\frac{1}{\pi D^2}\right) ln\left(\frac{\pi^2}{16} - \frac{1}{9}\left(\frac{\pi^2}{16}\right)^9\right) \quad (9)$$

$$D = 0.049\left(\frac{t_{0.5}}{l^2}\right)^{-1} \quad (10)$$

The approximate methods referred to as the slope method (Eq.8) and half-time method (Eq. 10) together with the full-form solution (Eq. 6) were used to estimate moisture diffusion of asphalt mastics in this paper. The plots from the slope method were also used to verify whether diffusion in asphalt mastics is Fickian in nature as discussed next.

Verification of Fickian diffusion

In the preceding sections on diffusion coefficients, a basic assumption was that moisture diffusion in asphalt mastics is Fickian in nature. Therefore, it was necessary to evaluate this assumption using the measured sorption data obtained in this study in order to verify the assumption of Fickian diffusion. Moisture diffusion in a material can be considered as Fickian if the kinetics of the moisture uptake satisfies certain important requirements (Comyn 1983, Crank 1975). First, a plot of $\frac{M_t}{M_\infty}$ against $\sqrt{t}$, should yield a straight line initially, followed by a curve that is concave with the time axis. Similarly, uptake curves of $\frac{M_t}{M_\infty}$ against $\frac{\sqrt{t}}{l}$ (i.e. reduced sorption curves) should coincide, approximately, regardless of film thickness. Finally, plots of uptake were evaluated to investigate the concentration dependence of asphalt mastic diffusion coefficient.

Materials and Methods

Materials

Mastic components

Limestone and granite aggregates were obtained from UK quarries. Previous studies (Airey et al., 2007) suggest these aggregates have significantly different susceptibility to moisture-induced damage under laboratory conditions. Therefore, it was expected that mastics made from the selected aggregates would show different sorption and diffusion characteristics. The aggregates were mechanically sieved in the laboratory to obtain only materials passing the 1-mm sieve and retained on 0.125-mm sieves (fine aggregate). The fine aggregate specific gravity values were 2.640 and 2.626, respectively, for the limestone and granite samples used (EN 12697, AASHTO T 84) which suggest the two aggregate types were similar in terms of density. Moisture absorption (EN 12697, AASHTO T84) for the limestone aggregates (0.70

213 percent) was relatively lower in magnitude than the granite (0.92 percent). The fine aggregate specific
214 surface areas, determined using octane probe and a dynamic sorption device, were 2.57 m^2/g and 3.49 m^2/g
215 for the limestone and granite, respectively.

216 In addition to the fine aggregates, limestone and granite mineral fillers satisfying BS EN 1097-7-
217 2008 were used. Again the choice of the mineral fillers was done to quantify the effect if any, of different
218 types of fillers used in asphalt mixtures.

219 A 40/60 penetration grade bitumen from a single source was used for preparing all the mastics. The
220 use of a single binder type was justified based on data from previous studies on moisture-induced damage
221 (Curtis et al. 1993) that suggested that aggregate chemistry is more influential than the composition of
222 asphalt binders. Bituminous binders similar to the ones used in this study are the single most common
223 binders used in the UK for asphalt concrete production and have been used to successfully study moisture-
224 induced damage in the past. Since moisture diffusion in a material is mainly influenced, at the molecular
225 level, by the physico-chemical properties of its constituents, it was expected that similar trends in moisture
226 diffusion behaviour would be obtained for different binder/aggregate combinations.

227 *Mastic Mix Design*

228 The proportion of the constituent components (fine aggregate, mineral filler, bitumen) of the
229 mastics was 50:25:25 by weight of mixture (Kringos et al. 2008). The aggregate components were weighed
230 separately, combined and thoroughly mixed in a mixing bucket, and heated at the mixing temperature of
231 185°C for about 10 minutes before adding hot bitumen (185°C) to the mixture. This relatively high mixing
232 temperature was used because of the high amount (25% w/w) of mineral filler used that results in very
233 viscous mixtures. The mixture was mixed using a Hobart mechanical mixer for about five minutes to
234 produce a homogenous mastic samples. The mastic were put in quart tins and stored in temperature
235 controlled (20°C, 50% RH) conditions until testing.

236 *Sorption Specimens*

237 Cylindrical disk specimens with nominal diameters of about 25 mm were used for evaluating the
238 sorption and diffusion characteristics of the mastics. They were prepared by carefully pouring molten
239 mastic (140°C) into specially designed silicone molds to form virtually void free specimens without any
240 compactive effort been applied. The average bulk specific gravity of the mastic measured using AASHTO

T166 was 1.917 (compared with a theoretically computed value of 1.907) which support the voidless (0% air voids) assumption. The thickness of the specimens ranged from about 1.50 mm to about 5.50 mm. The dimensions used in this study encompassed those used in previous studies. The specimens were cooled to room temperature and stored in a desiccator (to keep dry) until testing.

Table 2 shows details of the experimental matrix used for the study including the type of tests conducted, the type of mastics tested and the number of samples tested. Overall more than 30 individual mastic specimens were fabricated and tested.

Methods

Kinetic Vapor Sorption Profiles

Kinetic vapor sorption profiles for asphalt mastic were obtained using two different moisture vapor gravimetric sorption techniques. The first method involved the use of saturated potassium chloride salt solution at 23°C to generate a relative humidity of approximately 85% in a desiccator jar and manually measuring the weight gain with time (desiccator method). The weight gain (moisture uptake) of the mastic specimens was measured periodically (daily) using a Precisa XR 305A balance (Precision Balance Services Ltd) with a 0.1µg resolution. The moisture uptake was monitored until 'thermodynamic equilibrium' (i.e. a change of 0.0001g over a 24 hour period) for the conditions in the desiccator jar. At least three mastic specimens of each mix type were tested.

Some drawbacks of the desiccator method include the frequent (daily) opening and closing of the container (with attendant loss of vapor pressure) and the labor intensive nature of the method. Also, because of the stagnant nature of airflow within the desiccator jar, the time required to reach equilibrium is high (1-3 weeks). Despite these shortcomings, the desiccator method is easy to use, relatively cheap to setup, and several replicate specimens could be tested at the same time.

The second gravimetric method used was a dynamic sorption one and involved the use of an environmental chamber to precisely generate and maintain 85% RH at a temperature of 23°C while continuously measuring the weight gain of the mastic samples as it adsorbs water vapor (environmental chamber method). A TAS Model 600FS LTCL Series 3 environmental chamber capable of maintaining temperatures ranging from -75°C to 180°C and relative humidity from 0 to 95% was used. Air flow in the chamber was estimated to be about 0.50 m/s (about 0.10 m/s near the specimen). A Vibra HT-230CE tuning

fork analytical balance with a resolution of 0.1µg with static resistant plastic shield (to shield the specimen from the high speed air flow in the environmental chamber) was used to continuously monitor moisture uptake. Weight gain data was stored automatically via an RS-232C & peripheral device output to a personal computer. During the test, weight gain was captured every minute for a period of about 200 hours until 'thermodynamic equilibrium'. Compared to the desiccator method, the time to reach thermodynamic equilibrium was comparatively lower. The main advantage of the automated method includes the ability to dynamically monitor moisture uptake with higher accuracy in sorption profiles. The major drawback of the environmental chamber method is that only one sample could be tested at a time and it is relatively more expensive than the desiccator method.

Results and Discussion

Moisture Uptake Profiles

Moisture uptake profiles were computed as the ratio of moisture uptake at a given time to the original weight of the sample at the beginning of the test (Eq. 1). Sample moisture uptake results are presented in Figure 1 for the desiccator method and in Figure 2 for the climatic method. In both cases, moisture uptake increases rapidly at the beginning of the test and slows as "equilibrium" is approached. From Figure 1, it can be seen that the amount of moisture absorbed by asphalt mastic appears to be dependent on both thickness and aggregate type. Also the results suggest moisture uptake by mastic samples containing granite are relatively higher compared to limestone mixtures in most cases.

Similar to the results obtained for the desiccator method, differences in moisture uptake were seen based on the aggregate type in the climate chamber method. Mastic containing granite aggregate (GA) and / or granite filler (GF) exhibited moisture uptake profiles that plotted relatively higher than those containing limestone aggregates (Fig. 2).

Thus the trend in moisture uptake obtained from the desiccator method agrees with that obtained from the more accurate climate chamber method. However, the time to equilibrium moisture uptake appears to be shorter in the climatic chamber compared with the desiccator jar. The faster moisture uptake in the climate chamber could be attributed to several reasons related to the test set-up. First, the climate chamber method

297 is automated in terms of humidity generation and control. Secondly, the daily removal of specimens from
298 the sealed desiccators for weighing affects the partial vapor pressure generated by the saturated KCl solution
299 contained in the desiccator jar. Furthermore, unlike the desiccator method where the air around the sample
300 is static, in the climate chamber, air was constantly circulated by a high powered electric fan (air flow speed
301 = 0.5 m/s in the chamber).

302 **Equilibrium Moisture Uptake**

303 Equilibrium moisture uptake was computed as the maximum moisture uptake observed during a sorption
304 test (maximum uptake from Figs. 1 and 2). The exponential nature of Eq. 6 suggests that an infinite amount
305 of time is required for a sample to reach equilibrium moisture content. For practical reasons and since the
306 sensitivity of the balance used in this study was 0.0001 g, it was assumed that a sample reaches
307 "pseudoequilibrium" when the change in moisture uptake over a given 24 hour period does not exceed
308 0.0002 g. The equilibrium moisture uptake for the different mastics for both the desiccator method and the
309 climate chamber method are presented in Figure 3. Each value of moisture uptake in Fig. 3 is the arithmetic
310 mean for three or more samples of each mastic type, measured under the same experimental conditions.

311 In both gravimetric methods (Fig 3a and 3b), the magnitude of equilibrium moisture uptake appears
312 to be a function of aggregate type. Equilibrium moisture uptake was higher in the mastics containing granite
313 aggregate than in those with limestone. Several reasons could be used to explain the relatively higher
314 moisture absorption in the granite mastics. The results, showing that asphalt mastics containing granite
315 aggregates can absorb more moisture than the mastic containing limestone, appear to be in agreement with
316 the moisture absorption (obtained using AASHTO T 84) data previous presented for granite (0.9%) and
317 limestone (0.7%) fine aggregates. Another possible explanation for the higher moisture uptake in the granite
318 mastic could be attributed to the mineralogy of the aggregate. Granite is composed predominantly (about
319 72%) of silicon dioxide – a material with strong affinity for water and sometimes used as a desiccant – and
320 also alumina (about 14%) which previous studies (Fisher et al., 1922) suggest could absorb up to 18% of
321 its weight in moisture. The differences in the microstructure of the aggregates used could be another
322 possible reason for the differences in equilibrium moisture uptake obtained for the different asphalt mastics.
323 As previously mentioned, specific surface energy of the granite fine aggregate used in this study was
324 relatively higher than the limestone fine aggregate (3.49 m^2/g versus 2.57 m^2/g). Theoretically, the higher

the surface area of a material, the greater the potential sites for moisture vapor molecules to adhere to. The results of the current study as well those from previous studies (Fisher et al., 1922) suggest the asphalt mastic moisture uptake profiles (moisture diffusion and transport) depend on both mineralogical as well as microstructural characteristics of the constituent aggregates used. Therefore, both mineralogical and microstructural characteristics of mineral aggregates should have an important effect on moisture-damage susceptibility of asphalt mixtures.

Verification of Fickian diffusion

Asphalt mastic diffusion characteristics presented in this paper were based on the assumption that Fickian diffusion is applicable. It was therefore, necessary to verify the assumptions to assure the validity of the results as discussed next. Moisture diffusion in a material can be considered as Fickian if the kinetics of the moisture uptake satisfies certain important requirements (Comyn 1983, Crank, 1975).

First, a plot of $\frac{M_t}{M_\infty}$ against $\sqrt{t}$, should yield a straight line initially, followed by a curve that is concave with the time axis. Figure 4 depicts sample plots obtained for three mastics containing limestone aggregate and limestone filler showing linear relationship between $\frac{M_t}{M_\infty}$ and $\sqrt{t}$ that suggest diffusion in asphalt mastic might be Fickian in nature. Similar relationships were obtained for the other mixtures considered.

Determination of Diffusion Coefficient

Diffusion coefficient for the various mastics was determined using both the full-form solution and the simplified solutions. Determination of diffusion coefficient using the full-form solution utilized Eq. 6 and a numerical optimization routine by minimizing the sum of squares between the predicted moisture uptake and the measured uptake. For the simplified methods, Eq. 8 and Eq. 10 for the slope method and half-time method, respectively, were used to estimate diffusion coefficient of the mastics. It was observed (Figure 5) that both the simplified methods and the full-form solution of Fick's second law gave essentially the same results. The results suggest the two methods, full solution and half-time method could be used interchangeably to estimate diffusion coefficient of asphalt mastics.

Table 3 lists a summary of the average mastic moisture diffusion coefficients grouped in terms of mastic type, specimen thickness, equilibrium moisture uptake, and gravimetric sorption technique. The effects that these factors may have on mastic moisture diffusion coefficient are discussed next.

Factors influencing diffusion coefficient of asphalt mastics

One objective of this study was to identify factors that influence moisture diffusion in asphalt mastics in order to better understand moisture damage in asphalt mixtures. Four factors were considered including aggregate type, mineral filler type, specimen thickness, and moisture uptake (concentration). The importance of the first two factors (aggregate type and filler type) on moisture diffusion is obvious even though their effect on the actual mechanism of moisture damage is not clear. The third factor was important to consider because the diffusion coefficient determined in this study is expected to be used as input for numerical simulation of moisture damage in asphalt pavements and therefore any thickness effect (anisotropy), if any, needs to be documented. It is generally assumed that diffusion coefficient is an intrinsic material property that is independent of thickness but this has not been verified for asphalt mastics in previous studies. One assumption of Fickian diffusion is that diffusion coefficient is concentration dependent. Again this assumption has not been verified for asphalt mastics and therefore warrants this study.

Thickness effects

The results of asphalt mastic diffusion coefficients determination are presented in Figures 5 and 6 where a plot of diffusion coefficient against specimen thickness are compared for different combinations of aggregate and filler types.

The results suggest asphalt mastic diffusion coefficient increases with increase in thickness (Fig. 6) especially for the limestone aggregate mastic considered in this study. For specimen thickness less than 4.0 mm, moisture diffusion in limestone mastic appears to be similar to that of granite. Given that both mastics in Fig. 6 contain the same limestone filler (LF), the results suggest that for thinner specimens, the type of mineral filler may be a key factor influencing moisture diffusion. At greater thickness, limestone aggregate mastics moisture diffusion are relatively higher than the corresponding granite mastic which demonstrate the dominate influence of aggregate type on moisture transport. The effect of aggregate

mineralogy and microstructure could account for the higher rate of moisture transport (higher diffusion coefficient) in limestone mastics. As previously discussed, the limestone fractions considered in this paper have lower specific surface area (and hence lower moisture storage/adhesion capacity) than granites. It is also known that limestone aggregates have very different pore size and pore distribution (dual porosity) than granite. The finer pores in granite allows greater amounts of moisture to be adsorb and may have contributed to the observed differences in diffusivity observed in this study.

Mineral filler effects

For mastic containing the same aggregate type, the influence of mineral filler type on moisture diffusion appears to be minimal (Fig. 7). It must be noted that the amount of filler used was half that of the aggregate used and thus could have a limiting influence on moisture transport. Since replacing mineral filler with an active filler (such as hydrated lime) is one proven way of improving moisture sensitivity of asphalt mixtures, the minimal effect of filler on mastic diffusion is unexpected and warrants additional studies to investigate the phenomenon.

Longitudinal and radial diffusion

As previously discussed, the apparent relationship between asphalt mastic diffusion coefficient and specimen thickness was unexpected. One reason could be that both radial and longitudinal (through the thickness) diffusion could be occurring at the same time due to factors such as anisotropy (caused by aggregate settling to the bottom of the specimen during the long duration testing) and the violation of one of the assumptions (thin samples) of Fickian diffusion. For a material undergoing both radial and longitudinal diffusion (i.e. where radial diffusion may not be negligible), Eq. 11 (Crank, 1975) may be applicable and therefore, a linear relationship may exist between effective diffusion coefficient D (measured) and the square of specimen thickness ($\boldsymbol{l}$). Eq. 11 can therefore be used to 1) verify the existence of radial diffusion and 2) to estimate both radial and longitudinal diffusion coefficient components from the experimentally measured diffusion coefficient.

$$D = D_z + \left(\frac{2D_r}{\pi^2 a^2}\right) l^2 \qquad (11)$$

where D_r = the radial or lateral diffusion coefficient

D_z = the longitudinal diffusion coefficient

a = radius of specimen

As shown in Figure 8, the relationship between D and $\boldsymbol{l^2}$ is approximately linear for most of the mastics tested. This demonstrates that both radial and longitudinal diffusion might be occurring in the asphalt mastics tested, especially in thicker specimens. The results (slope and intercept values) from Figure 8 were used to estimate radial and longitudinal diffusivity of asphalt mastic as presented in Table 4. Because of the limited number of specimens tested, the results should be considered as tentative. It is interesting, however, to note the close agreement between the longitudinal diffusion coefficient obtained in this study (0.10 to 1.23 x 10^{-12} m^2/s) and the corrected (both experimentally and numerically) diffusion coefficient values reported for similar mastics (approximately 1 mm thick) by Kringos et al. 2008. It is also interesting to note the similarity (in terms of order of magnitude) between radial diffusion coefficient (65 to 362 x 10^{-12} m^2/s) obtained in this study and reported aggregate diffusion coefficient in previous studies (122 to 200 x 10^{-12} m^2/s by Kringos et al. 2008 and 210 to 245 10^{-12} m^2/s reported by Arambula et al. 2010) which would suggest the radial diffusion observed in the current study may be due to aggregate particles that settles in the mastic as previously discussed.

Thickness-dependent diffusivity has not previously reported in asphalt mastics even though the phenomenon has been observed in other materials (Fernando et al., 2011; Pereira and Yarwood, 1996; Tutuncu and Labuza, 1996). Additional studies looking at radial and longitudinal diffusion in asphalt mastics is warranted. However, the limited data obtained in this study suggest anisotropic diffusivity in asphalt mastics. For homogenous and isotropic materials, diffusivity can be considered as an intrinsic property that is independent of size (sample thickness). The observed variation of diffusivity with thickness (resulting in apparent radial and longitudinal diffusivities) reported in this paper suggests asphalt mastic may not be isotropic and/or homogenous. The authors believe that asphalt mastic, under the laboratory conditions used during a typical gravimetric moisture diffusion experiment (isothermal condition, tests lasting 100s of hours), could result in significant settlement of the denser fine aggregate particles to the bottom of the sample leaving the lighter mineral filler components near the upper portions of the sample. It is conceivable that the settling aggregates could in turn result in pockets of minute cavities (air pockets) at

435 the original locations of the aggregate. The probability of these effects occurring would obviously increase
436 with sample thickness. The consequence of these two changes is a mastic sample that can be considered as
437 anisotropic (i.e. a graded material with most of the more porous aggregates settled at the bottom and lighter
438 but less porous material near the top). These are further compounded by the fact that diffusion coefficient
439 of aggregates (122 to 200 x 10^{-12} m^2/s [Kringos et al. 2008]; 80800 x 10^{-12} m^2/s [Henon et al. 2002]) and air
440 (26000000 x 10^{-12} m^2/s) are significantly larger than that of asphalt binders and therefore any small changes
441 in their amount could large effect on diffusivity of mastics.
442
443 ***Concentration-dependent diffusion coefficient***
444 The concentration dependence of moisture diffusion in asphalt mastic was analyzed by plotting diffusion
445 coefficient against equilibrium moisture uptake for the four different mastics considered in the study (Fig.
446 9). In all cases, diffusion coefficient appears to decrease with increasing equilibrium moisture content. The
447 relationship between moisture absorption and diffusion coefficient of asphalt mastics can thus be
448 characterized by an inversely proportional function of moisture uptake and diffusivity. These results
449 showing concentration-dependent coefficient of diffusion suggest moisture transport in asphalt mastics is a
450 function of aggregate type and that both equilibrium moisture uptake and diffusion coefficient should be
451 considered when characterizing moisture transport in asphalt mixtures. Some previous studies (Kringos et
452 al. 2008; Chen et al. 2003) have suggested, based on the higher equilibrium moisture uptake and lower
453 diffusion coefficient of granites, that equilibrium moisture uptake is more useful than the magnitude of
454 diffusion coefficient for characterizing moisture susceptibility. The results of the current study showing
455 concentration dependent diffusion coefficient suggest both parameters are complimentary and should be
456 considered together when evaluating moisture diffusion in asphalt mixtures.
457
458 ***Simulation of diffusion in asphalt mastics***
459 Moisture diffusion in asphalt mastics can be simulated using values of diffusion coefficient and equilibrium
460 moisture uptake obtained in this study. Figure 10 and Figure 11 show sample plots of moisture diffusion in
461 two of the mastic studied. A 4-mm thick specimen was assumed for both mastics. Diffusion coefficient was
462 taken as 2.043x10^{-12} m^2/s and 1.697x10^{-12} m^2/s for mastics containing limestone (LA+LF) and granite

(GA+GF) mastic, respectively. The corresponding equilibrium moisture uptakes were 0.0994% and 0.1459%, respectively for limestone and granite mastics. The plot illustrates differences in the rate as well as magnitude of equilibrium moisture uptake in mastics. For example, the plots show that after about 100 hours and at a depth of 0.5 mm from the surface of the mastic specimen, about 0.068% of moisture had been absorbed in the limestone mastic compared to about 0.095% in the granite. Given that each of the mastic types illustrated in Figures 10 and 11 contained the same aggregate types, the results suggest it takes the granite mastic longer to reach equilibrium moisture content compared with limestone aggregate. The results appear to agree with previously reported values for moisture diffusion in asphalt mastic (Kringos et al. 2008).

Summary and Conclusions

This study was conducted to characterize the sorption and diffusion characteristics of asphalt mastic using gravimetric vapor sorption techniques. Moisture vapor sorption tests were conducted at 23°C and 85% RH on four asphalt mastics fabricated using the same pen 40/60 bitumen but two different aggregates and two different mineral filler types. The following conclusions were reached based on the results of the study.

- The kinetics of moisture uptake obtained was characteristic of Fickian diffusion with a concentration-dependent diffusion coefficient.
- Equilibrium moisture uptake and diffusion coefficient estimated from the static measurements were comparable and of the same order of magnitude as those from dynamic sorption techniques. Both measurement techniques ranked the mixes similarly which suggest either method could be used to characterize moisture transport in asphalt mastics.
- Equilibrium moisture uptake was relatively higher in mixtures containing granite aggregates compared with limestone aggregate. In contrast, the diffusion coefficient of limestone aggregate mastics was higher than granite. Thus the relationship between moisture absorption and diffusion coefficient of asphalt mastics can be characterized by an inversely proportional function of moisture uptake and diffusivity. The results suggest moisture transport is a function of aggregate

type and that both equilibrium moisture uptake and diffusion coefficient are useful in studying moisture susceptibility in asphalt mixtures.

- The effect of mineral filler type on diffusion coefficient was minimal in the mastics containing granite aggregate but relatively high in mastic samples containing limestone aggregates.
- Diffusion coefficient was found to increase with sample thickness which was unexpected because diffusion coefficient (in an isotropic material) is considered an intrinsic property that is independent on sample size. The results suggested anisotropic diffusivity can occur in asphalt mastics and could be attributed to factors including mineralogy, microstructure, air voids, and the tendency of the aggregates to settle at the bottom of asphalt mastic with time.
- In addition to characterizing moisture transport in asphalt mastics, the results presented in this paper will be useful as inputs for numerical simulation of moisture damage in asphalt mixtures.

Acknowledgements

The authors acknowledge the assistance of the following Nottingham Transportation Engineering Centre personnel: Martyn Barrett, Richard Blakemore, Nancy Milne, Lawrence Pont, Jonathan Watson, and Michael Winfield for material testing. The funding for this project was provided in part by the UK Engineering and Physical Sciences Research Council (EPSRC).

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558 **List of Tables**

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563 Table 1. Sample reported asphalt mastic diffusion coefficients

Reference	Blend proportions (by weight)	D x 10^{-12} (m^2/s)	Specimen characteristics and experimental conditions
Kassem et al. (2006)	Aggregate: 52.4% sandstone, 35.0% igneous screening, 4.6% hydrated lime asphalt: 8.0% PG 76-22	10.26	Cylindrical specimens, 50 mm in diameter and 50 mm in height, sitting in a shallow water bath at 25°C, while measuring the change in the logarithm of total suction using a psychrometer embedded in the middle specimen and placed 5 mm above the bottom end of the of the specimen. Aggregate size passing sieve Number 16 (1.18mm). Air voids not specified.
	Aggregate: 66.2% natural sand, 25.8% limestone sand asphalt: 8.0% (PG 64-22 & PG 64-28)	9.72 & 24.30	
Kringos et al. 2008	Aggregate: 50% crushed sand, 25% lime asphalt: 25% binder (Pen 70/100, Cariphalt XS, Sealoflex)	0.13 – 3.08	Gravimetric sorption method applied to 30 mm × 30 mm and 1 mm thick specimens placed inside an 85% relative humidity chamber at 25°C. Aggregate size (top size 1.18 mm). Air voids not specified.
Arambula et al. (2010)	Aggregate: 47.3% diabase, 42.5% sand, 1.7% dust asphalt: 8.5% PG 70-22	25444	Gravimetric method (ASTM E96, wet cup/dry cup method) applied to cylindrical ensembles containing 70 mm diameter and 4–5 mm thick specimens, where the ensembles were placed in a chamber with 15% relative humidity at 35°C. Aggregate size passing sieve No. 4 (4.75 mm). Air voids between 11–13%. Used Fick's first law to estimate D.
Vasconcelos et al. (2011)	Aggregate: N/A, asphalt: 8.9% (PG 58-XX, XX=10, 22, 28)	0.78 – 2.23	23.9 °C temp. Conditioning by submerging in water for 21 months and measuring SSD with time using a sensitive mass balance. Aggregate size passing sieve 1.18 mm sieve. Air voids between 7–11%. Specimen size 12 mm diamter by 50 mm thick.
	Aggregate: N/A asphalt: 8.9% (PG 58-XX, XX=10, 22, 28)	2.21 – 4.90	37.8 °C temp. Conditioning by submerging in water for 14 months and measuring SSD with time using a sensitive mass balance. Aggregate size passing sieve 1.18 mm sieve. Air voids between 7–11%. Specimen size 12 mm diamter by 50 mm thick.

564 N/A = data not available
565

566 Table 2. Experimental matrix

Matic type	Test method	No. specimens tested	[1]Thickness (mm)
LA + LF	Desiccator jar	8	3.19 ± 1.50
	Environmental chamber	3	3.37 ± 1.04
LA + GF	Desiccator jar	6	2.63 ± 1.08
	Environmental chamber	3	4.22 ± 0.50
GA + LF	Desiccator jar	3	3.93 ± 1.09
	Environmental chamber	3	3.37 ± 0.27
GA + GF	Desiccator jar	3	4.32 ± 0.71
	Environmental chamber	3	3.53 ± 0.98

LA = Limestone aggregate, LF = limestone filler, GA = granite aggregate, GF = granite filler. All mastics contained 40/60 pen bitumen. [1] = Mean ± SD

567
568

569
570 Table 3. Moisture diffusion coefficient of asphalt mastics at 23°C based on desiccator and climatic chamber methods.

Matic type	Test method	No. specimens tested	[1]Thickness (mm)	Equilibrium moisture uptake (%)	Diffusion coefficient x 10^{-12} (m^2/s)
LA + LF	Desiccator jar	8	3.19 ± 1.50	0.1136 ± 0.025	4.02 ± 5.09
	Climatic chamber	3	3.37 ± 1.04	0.0738 ± 0.023	2.59 ± 0.22
LA + GF	Desiccator jar	6	2.63 ± 1.08	0.1236 ± 0.023	1.33 ± 0.74
	Climatic chamber	3	4.22 ± 0.50	0.0811 ± 0.006	4.75 ± 0.06
GA + LF	Desiccator jar	3	3.93 ± 1.09	0.1211 ± 0.032	2.04 ± 1.07
	Climatic chamber	3	3.37 ± 0.27	0.1095 ± 0.001	2.87 ± 0.01
GA + GF	Desiccator jar	3	4.32 ± 0.71	0.1198 ± 0.022	2.17 ± 0.48
	Climatic chamber	3	3.53 ± 0.98	0.1484 ± 0.022	2.44 ± 0.22

LA = Limestone aggregate, LF = limestone filler, GA = granite aggregate, GF = granite filler. All mastics contained 40/60 pen bitumen. [1] = Mean ± standard deviation

571

572 Table 4. Longitudinal and radial diffusion coefficient of asphalt mastic

Matic type	Test method	No. specimens tested	[1]Thickness (mm)	Long Diffusion coefficient x 10^{-12} (m^2/s)	Radial Diffusion coefficient x 10^{-12} (m^2/s)
LA + LF	Desiccator jar	8	3.19 ± 1.50	1.23 ± 1.61	361.57 ± 87.18
	Climatic chamber	3	3.37 ± 1.04	0.09 ± 0.23	173.12 ± 14.00
LA + GF	Desiccator jar	6	2.63 ± 1.08	0.37 ± 0.20	101.99 ± 17.91
	Climatic chamber	3	4.22 ± 0.50	0.48 ± 0.55	206.72 ± 25.71
GA + LF	Desiccator jar	3	3.93 ± 1.09	0.18 ± 0.26	95.66 ± 12.01
	Climatic chamber	3	3.37 ± 0.27	0.45 ± 2.19	242.18 ± 159.08
GA + GF	Desiccator jar	3	4.32 ± 0.71	0.70 ± 0.53	64.84 ± 22.64
	Climatic chamber	3	3.53 ± 0.98	0.51 ± 0.06	122.92 ± 3.69

LA = Limestone aggregate, LF = limestone filler, GA = granite aggregate, GF = granite filler. All mastics contained 40/60 pen bitumen. [1] = Mean ± standard deviation

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FIGURES

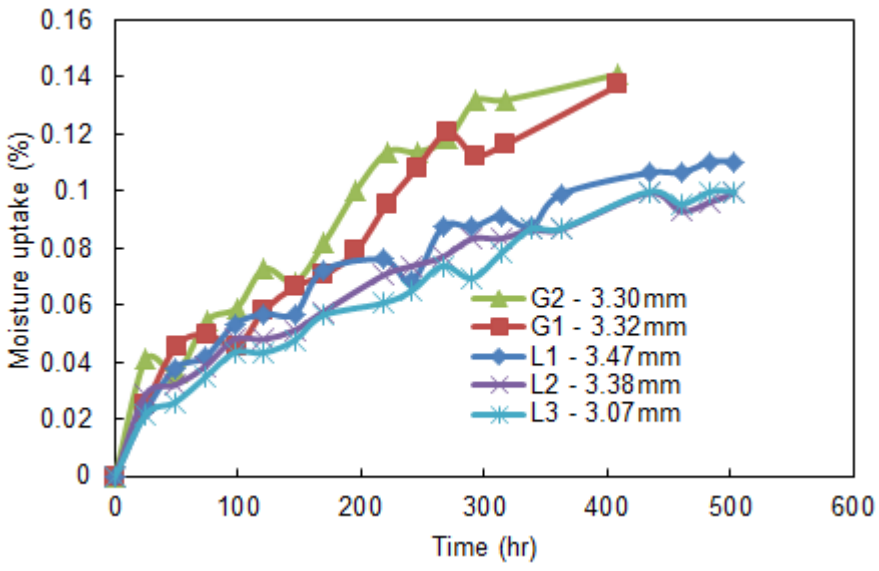


Fig. 1. Sample asphalt mastic moisture uptake profiles obtained using desiccator method. Moisture uptake for mastics containing granite aggregate and filler (G1 and G2) were relatively higher than limestone aggregate and filler (L1-L3) mastics. All specimens contained the same 40/60 pen asphalt binder and limestone mineral filler.

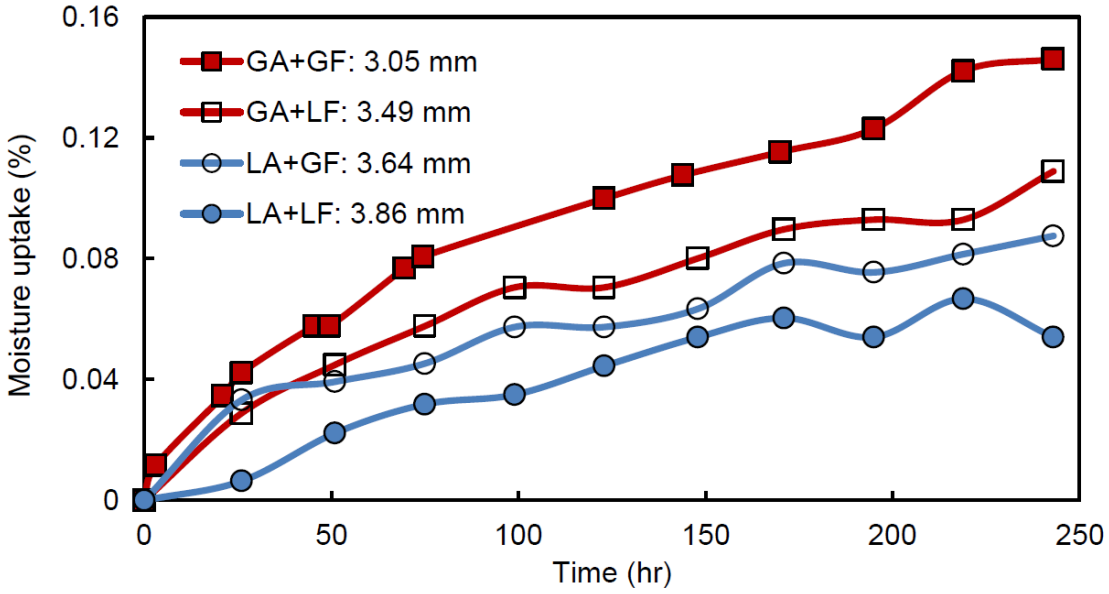


Fig. 2. Sample asphalt mastic moisture uptake profiles obtained using climate chamber method. Moisture uptake for mastics containing granite (GA and GF) plotted relatively higher than limestone (LA and LF) mastics which seems to agree quite well with the desiccator method.

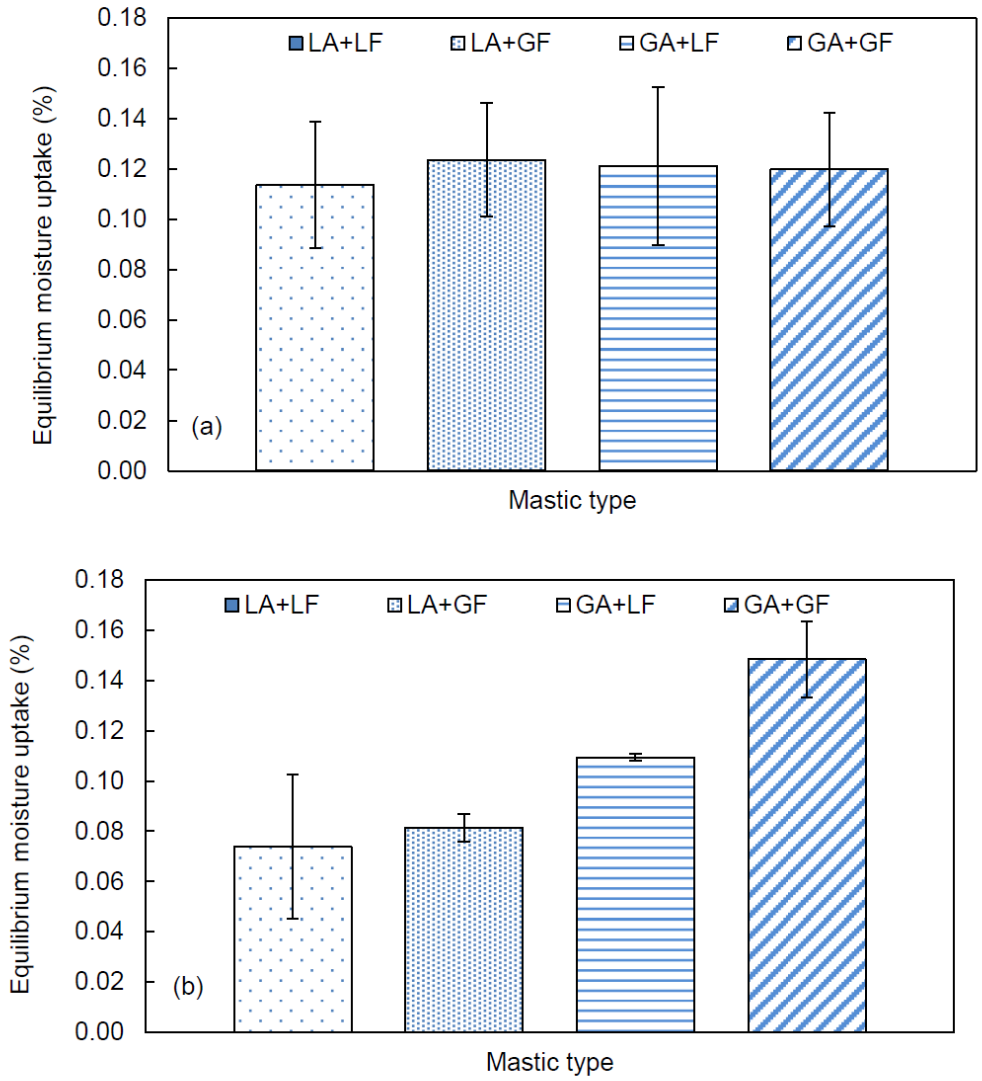


Fig. 3. Equilibrium moisture uptake for asphalt mastic obtained from desiccator jar (a) and climate chamber (b) methods. Each value of moisture uptake in Fig. 3 is the arithmetic mean for three or more samples of each mastic type, measured under the same experimental conditions. The error bars represent ± one standard deviation. LA+LF = mastic with limestone aggregate and limestone filler. LA+GF = mastic with limestone aggregate and granite filler. GA+LF = mastic with granite aggregate and limestone filler. GA+GF = granite aggregate with granite filler. All mastics contained 40/60 pen bitumen.

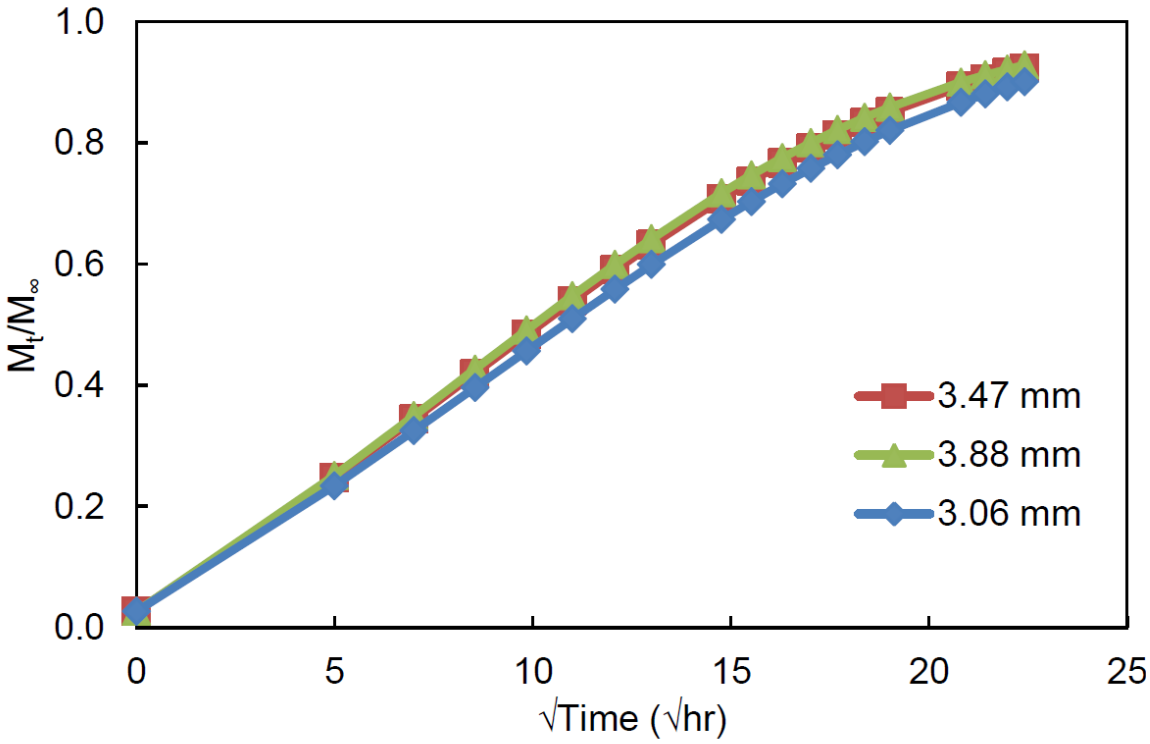


Fig. 4. Values of $\frac{M_t}{M_\infty}$ plotted against $\sqrt{t}$ for asphalt mastic gives a straight line for $\frac{M_t}{M_\infty} < 0.6$ suggesting Fickian diffusion. Values plotted were obtained from three replicates of the same mastic.

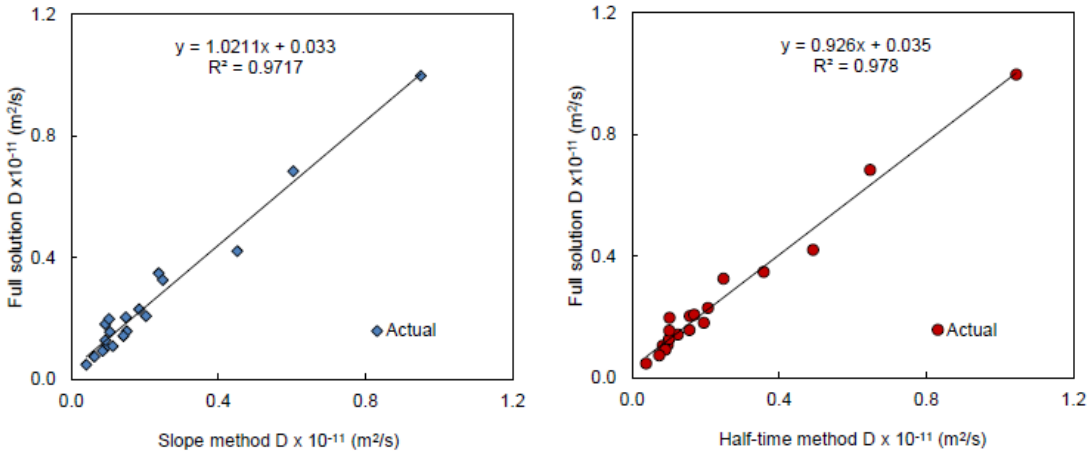
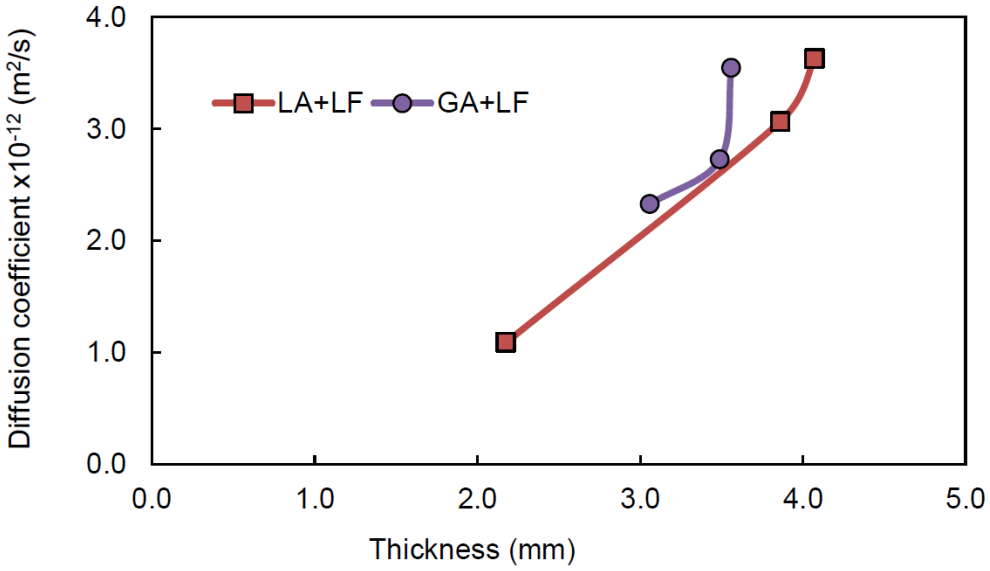


Fig. 5. Estimation of diffusion coefficient for asphalt mastic using simplified and full-term solutions. Half-time method agrees better with full-term solution than slope method. Data shown are for 19 different mastic specimens.



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638 Fig. 6. Effects of aggregate type and specimen thickness on moisture diffusion of asphalt mastics containing the
639 same limestone mineral filler (LF).

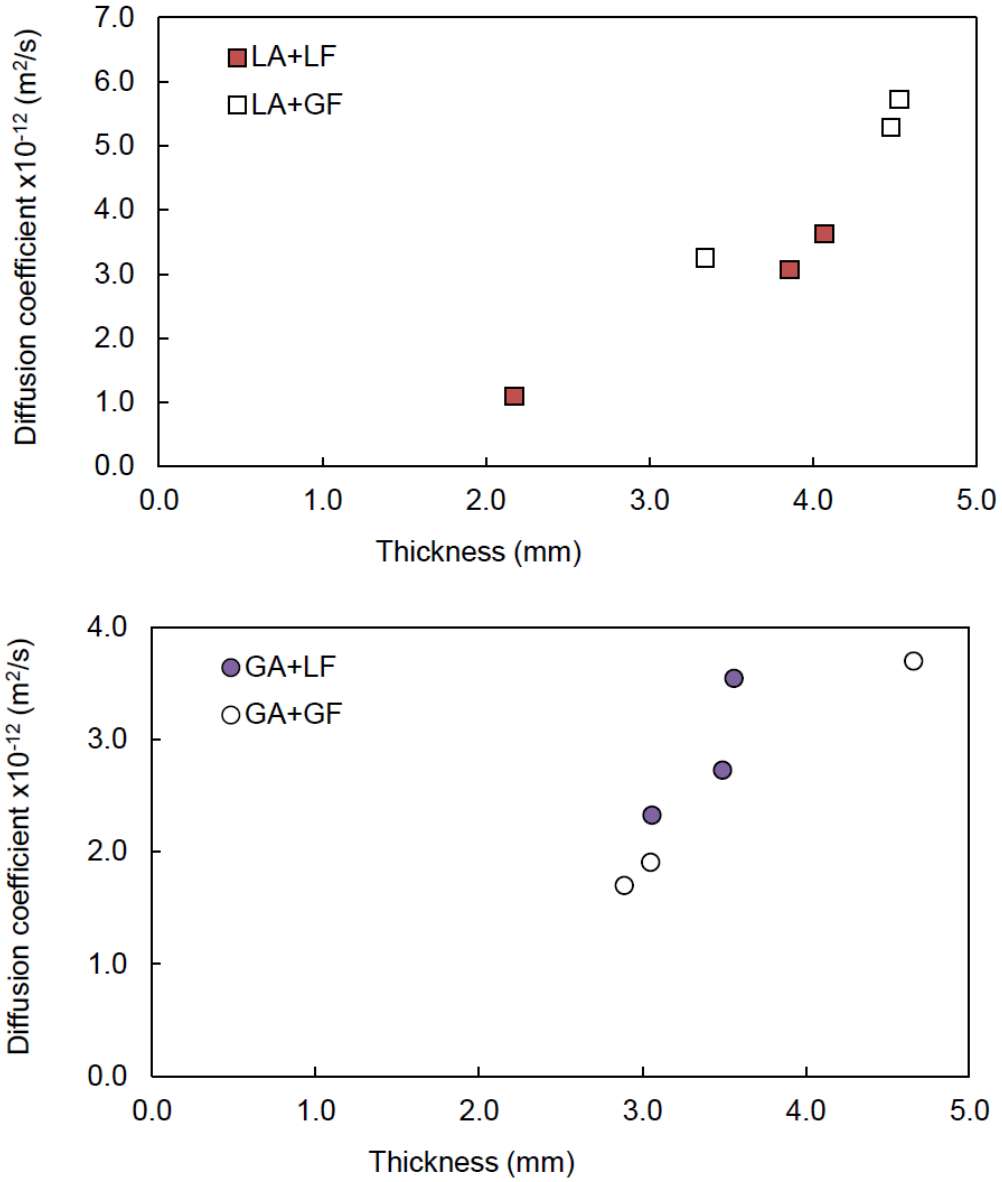

Fig. 7. Effects of mineral filler type on asphalt mastic diffusion coefficient. For the same aggregate type (limestone, LA; or granite, GA, neither the limestone filler (LF) nor the granite filler (GF) significantly altered moisture diffusion in asphalt mastic.

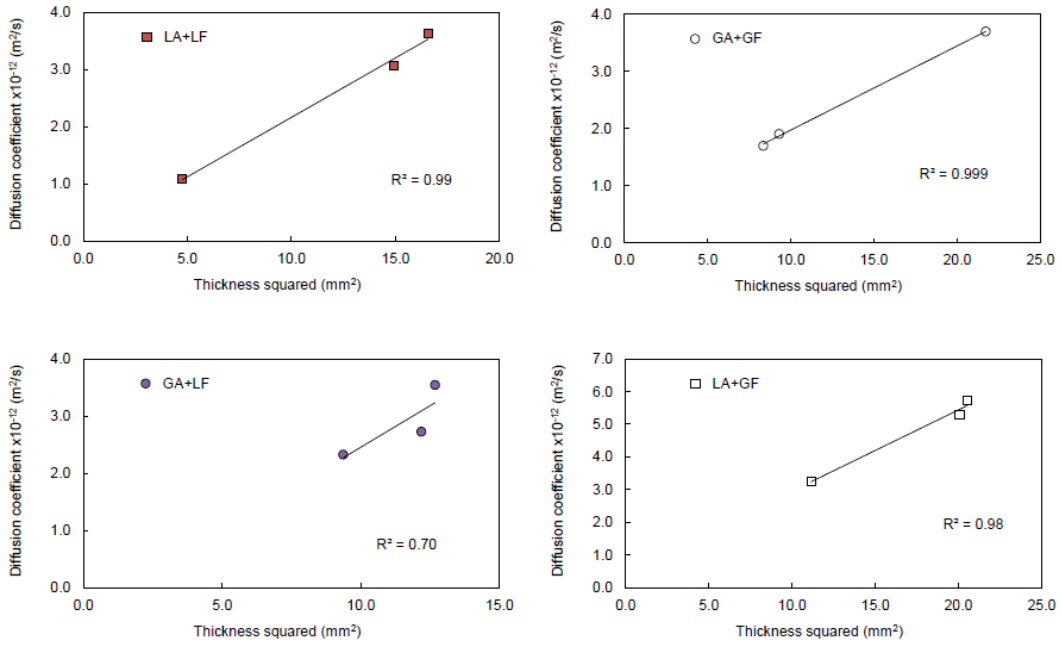

Fig. 8. Investigation of radial and longitudinal diffusion in asphalt mastic. Linear relationship between diffusion and thickness suggest both modes of diffusion occurs in mastics.

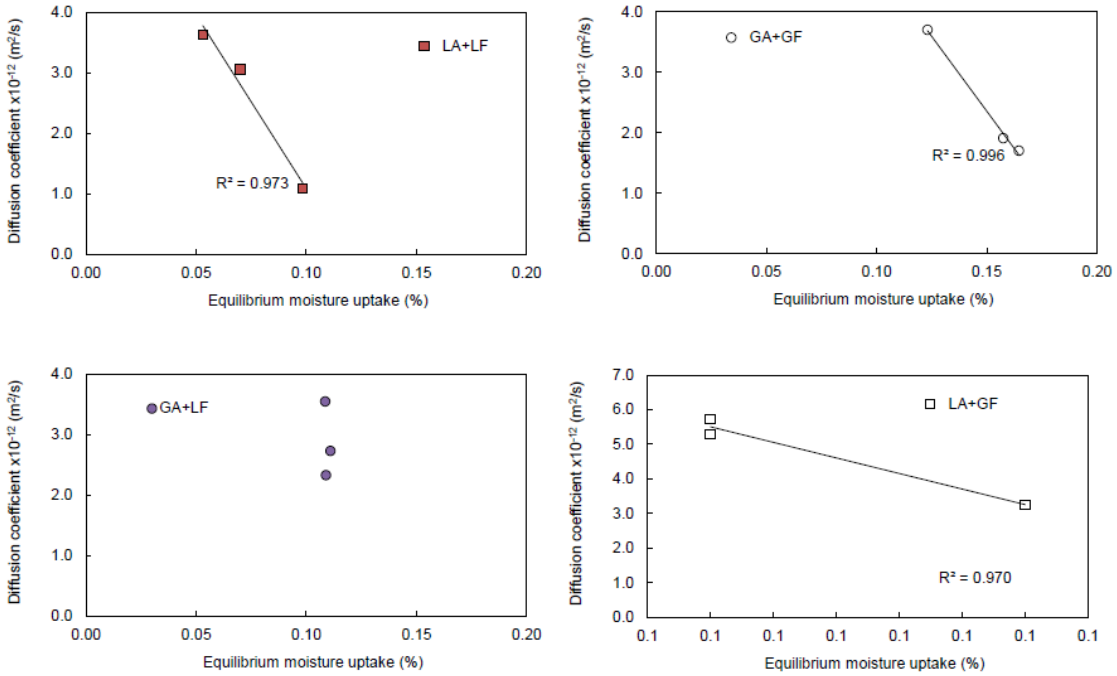

Fig. 9. Plots of diffusion coefficient against equilibrium moisture uptake showing linearly decreasing diffusivity with moisture content for four different asphalt mastic types. The results demonstrate moisture diffusion in asphalt mastic is concentration dependent.

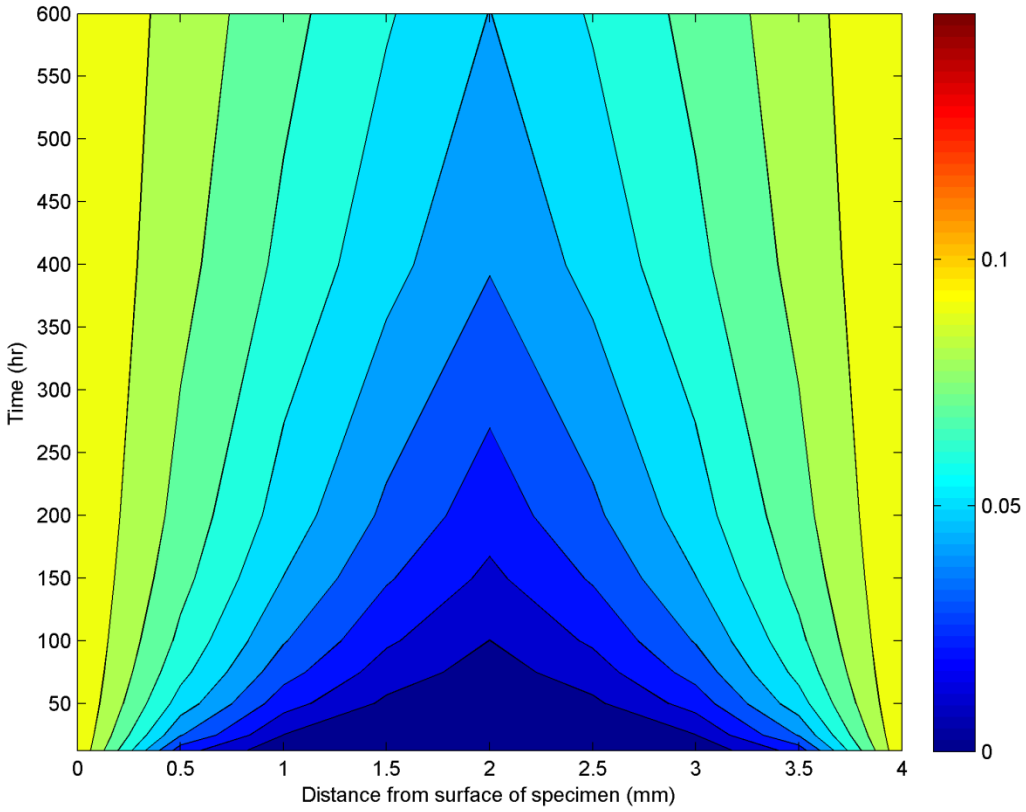

Fig. 10. Simulation of moisture diffusion in asphalt mastic containing limestone aggregate and limestone filler. Testing conditions simulated included 85% RH at a temperature of 23°C for moisture content ranging from about 0 to 0.10%. Plotted values are moisture uptake (%).

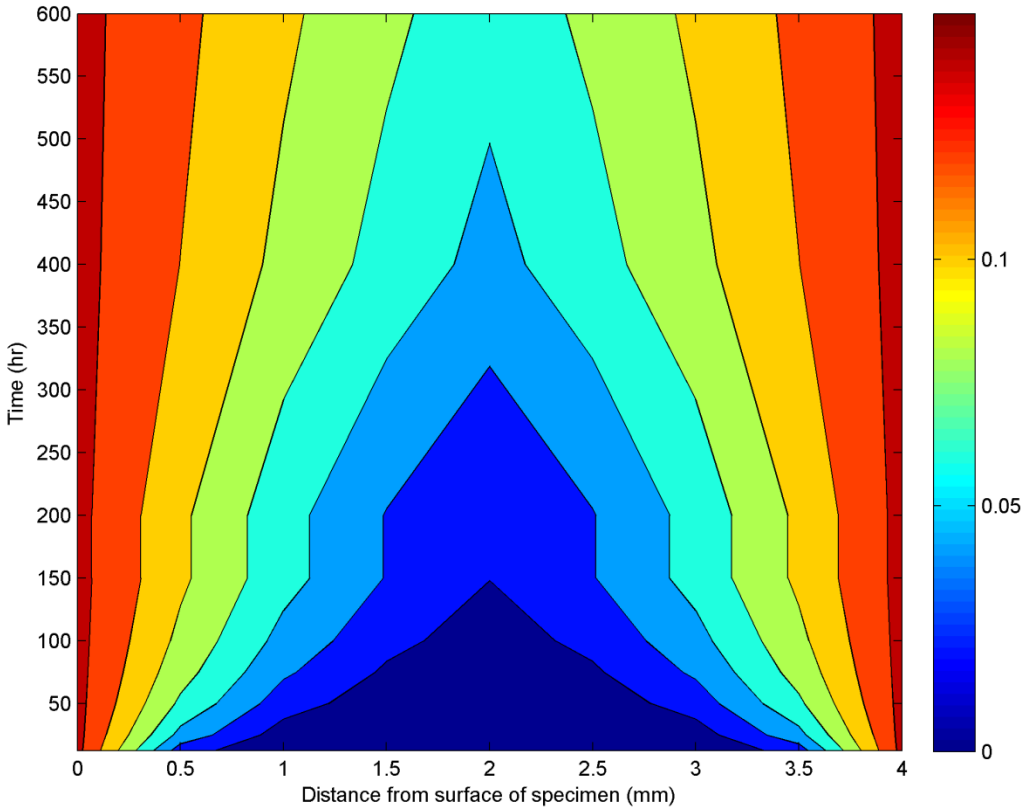

Fig. 11. Simulation of moisture diffusion in asphalt mastic containing granite aggregate and granite filler. Testing conditions simulated included 85% RH at a temperature of 23°C for moisture content ranging from about 0 to 0.15%. Plotted values are moisture uptake (%).