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Observation of reversible moisture damage in asphalt mixtures

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HIGHLIGHTS

• Durability of asphalt mixtures conditioned in water at 60 °C was investigated.

• Durability was evaluated using indirect tensile tests at 20 °C.

• Moisture conditioned asphalt mixtures lost up to 80% of the initial stiffness.

• Upon drying the mixtures fully recovered both their stiffness and tensile.

The results suggest moisture damage in the mixtures tested is reversible.

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1. Introduction

There is little doubt about the detrimental effects that the presence of moisture has on the extent and severity of most pavement distresses around the world [1]. In the UK alone, billions of pounds are spent annually to repair the damage to pavement caused in part by the effect of moisture on asphalt mixtures [2]. A major challenge is the lack of fundamental understanding of the mechanism by which the presence of moisture in an asphalt mixture leads to damage. As a result many empirical tests have been developed in the past to predict moisture sensitivity of asphalt mixtures [3]. Recent attempts aimed at characterizing moisture-induced

ABSTRACT

Durability of asphalt mixtures conditioned in hot water was investigated using stiffness measurements. Stiffness generally decreased with conditioning time. The effect of moisture on stiffness was found to be reversible as moisture conditioned-asphalt mixtures that had lost up to 80% of their initial stiffness fully recovered upon subsequent drying. Estimates of mastic film thickness and length of diffusion paths obtained from image analysis of X-ray CT scans of the asphalt mixtures suggest moisture diffusion was mainly restricted to the bulk mastic. The results suggest cohesive rather than adhesive failure dominated the durability of asphalt mixtures under the long-term moisture exposure used in this study. © 2014 The Authors. Published by Elsevier Ltd. This is an open access article under the CC BY license (http://

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damage in asphalt–aggregate mixtures in a more fundamental way have focused on applying physical adsorption theories [4]. The approach involves surface free energy measurements of the individual constituents of asphalt mixtures (aggregate and bitumen) by applying vapor sorption techniques and using the results to calculate thermodynamic work of adhesion and debonding in the presence of moisture, of various aggregate–asphalt mixtures. The physical adsorption approach represents a vast improvement over the existing empirical moisture sensitivity tests because it applies fundamental concepts of adhesion that is based on the molecular interaction between an adhesive and a substrate. Because the approach is based on fundamental material properties, the physical adsorption theory of moisture-induced damage is applicable to all mixture types unlike some current empirical tests that are both material and test method dependent.

The basic concepts behind physical adsorption theory suggest that (1) the adhesive and the substrate are in intimate contact

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and van der Waals forces operate between them, (2) van der Waals forces consist of two components – polar and dispersion, and (3) thermodynamic work of adhesion, calculated by using the twocomponent van der Waals forces, can be used to assess the stability of the bond between an adhesive and a substrate [5]. Adhesivesubstrate bonds with positive thermodynamic work of adhesion are considered stable while bonds with negative work of adhesion are considered unstable. Energy parameters based on thermodynamic work of adhesion for characterizing moisture sensitivity have been developed for asphalt mixtures [4].

One important limitation of the physical adsorption theorybased moisture damage evaluation technique is that even though the technique can predict the stability of an adhesive-substrate bond under both wet and dry conditions, it is unable to account for the moisture damage that is reversible upon specimen drying. This type of damage has been observed in asphalt mixtures in previous studies, even though the phenomenon has neither been reported widely nor fully explained. For instance, evidence of moisture-induced stiffness degradation in wet specimens being fully recovered upon specimen drying was reported by Schmidt and Graf [6]. The authors reported that resilient modulus of moisture-deteriorated specimens returned to their original value on drying and attributed the occurrence of the phenomenon to the observed disparity between lab test data and field performance. The lack of correlation between field performance and some laboratory moisture sensitivity tests could be due to the fact that existing test methods and pavement design analyses techniques do not account fully for the reversibility of the moisture-induced stiffness degradation phenomenon directly. The chemical bonding theory could be used to address reversible stiffness degradation in moisturedamaged mixtures. The formation of covalent, ionic, or hydrogen bonds across an adhesive-substrate interface is the basis for the chemical bonding theory of adhesion [5]. The interfacial force due to ionic pairs is given by Eq. (1) [7], where q_1 and q_2 are the ionic charges, ϵ_0 the permittivity of a vacuum, ϵ_r the relative permittivity of the medium, and *r* the inter-ionic distance.

$$F = \frac{q_1 q_2}{4\pi \varepsilon_0 \varepsilon_r r^2} \tag{1}$$

Water at ambient temperatures has a very high relative permittivity of 80. The corresponding relative permittivity of bituminous materials, however, are quite low with reported values in the range of 2.6–2.8 for bitumen, 4.0–4.6 for newly constructed dry asphalt pavements, and 6–8 for wet or moisture damaged pavements [8– 11]. Since an approximately linear relationship exists between the relative permittivities of mixtures of water and organic solvents and mixture composition [7,12,13], the high ε_r of water means even small amounts of absorbed water in the adhesive can cause large increases in ε_r and a reduction in F. For asphalt mixtures, assuming an increase in relative permeability from 4.3 for a new asphalt mixture to 7.0 for old/moisture damaged mixtures, a reduction in F of about 63% is possible. The reduction in F due to moisture absorption by an adhesive is reversible; hence complete removal of water (say by drying) from an adhesive joint can restore F to the original value. Thus the major difference between the adsorption theory and the chemical bond theory of adhesion is that the latter permits partial recovery of damage in a wet adhesive bond when the bond is dried while the former determines whether an adhesive bond is stable or not stable (zero strength) in the presence of water. The insight gained from the work presented in the current paper is intended to help to develop a framework for describing the mechanism responsible for the reversibility of moisture-induced stiffness degradation in asphalt mixtures and suggest a unified adhesion-cohesion theory to characterize moisture sensitivity of asphalt mixtures.

This paper presents indirect tensile testing results for densegraded asphalt mixtures that contain limestone aggregates and two different mineral fillers (granite and limestone) and that had been compacted to three different air void levels and subjected to water immersion testing at 60 °C for up to 70 days. The mechanical test data were compared with the thermodynamic work of adhesion of the asphalt mixture components. Moisture diffusion analysis and computerized tomography techniques were used to estimate moisture transport in the asphalts mixtures in order to determine the possibility of moisture reaching the aggregate-mastic interface. Adhesion theory, based on chemical (covalent bonds or ionic pairs) were used to explain the apparent recovery of stiffness degradation in previously moisture-damaged specimens. Analysis of the results lead to the development of a novel framework for describing moisture-induced damage in asphalt mixtures in terms of both adhesive and cohesive failure.

2. Materials and methods

2.1. Mixture composition, design and conditioning

The asphalt mixtures used were 10 mm nominal maximum size dense bitumen macadam (DBM) mixtures that had been compacted to three different air void levels using a gyratory compactor. The selection of the constituent aggregate, mineral filler, and bitumen were based on experience from previous studies that found mixtures fabricated from these materials exhibit different moisture sensitivity depending on aggregate and or mineral filler type [14,15]. For each aggregate, mineral filler, and air void level, four replicate 100 mm diameter by 60 mm thick specimens were fabricated for moisture conditioning and mechanical testing. Overall, 48 asphalt (100 mm by 60 mm) specimens were fabricated for moisture conditioning and mechanical testing (indirect tensile stiffness and tensile strength). Of these, 24 specimens were tested in the dry condition without moisture conditioning to determine their tensile strength while the rest were conditioned by immersion in a water bath at 60 °C for up to 70 days. The moisture-conditioned specimens were removed at regular intervals for testing and then returned to the water bath for additional conditioning. Additional details of the asphalt mixtures used, including the constituent material properties, key mixture design features, and moisture conditioning, are listed in Table 1. Fig. 1 shows a photograph of the moisture conditioning test set-up used for asphalt mixture immersion test.

2.2. Physico-chemical properties of aggregates and bitumen

Physico-chemical properties of the aggregate and bitumen used were determined by using vapor sorption techniques and contact angle measurements, respectively. Detailed experimental procedures used, including selection of probe liquids and procedures for estimating surface free energy components of the aggregate and bitumen, are provided elsewhere [16,17]. The data were used to calculate the thermodynamic work of adhesion and debonding of the aggregate–bitumen mixtures. Summary of the physico-chemical properties of the bitumen and the aggregate including the thermodynamic work of adhesion are listed in Table 2.

As expected the thermodynamic work of adhesion between the aggregate and bitumen in the dry state was positive suggesting a stable bond. However, the negative nature of the work of debonding suggests the bond between aggregate and bitumen in the presence of water is unstable, which is in agreement with expectation based on common experience. It is important to note in Table 2 that the work adhesion between the aggregate and the bitumen is higher than the cohesion within the bitumen. This would suggest that in the absence of water, the dominant failure mode in asphalt mixtures should be cohesive. This observation will be used to develop the moisture damage mechanism framework introduced in this paper.

2.3. Indirect tensile stiffness testing of mixtures

Indirect tensile stiffness tests were performed in accordance with EN 12697-26 [18] on the 100 mm diameter by 60 mm thick specimens at various moisture conditioning times starting from the dry condition until the end of a 70 day period. Specifically specimens were removed from the water bath and tested on the following days: 8, 15, 30, 55, and 70. In each instance, except for specimens conditional specimen conditioning. After the 70 day testing, the specimens were stored at room temperature (20 °C and 55% relative humidity) for another 47 days to dry and were then tested. The latter testing enabled a determination to be made as to whether moisture-induced stiffness degradation in asphalt mixtures was reversible. All the stiffness testing was conducted at 20 °C. The applied load was selected to ensure that the mixture deformation stayed in the linear viscoelastic range in order not to damage the specimens as the same specimens were tested and then put back in the water bath for additional conditioning.

Table 1	
Details of asphalt mixtures cons	stituents, key mix design parameters, and moisture conditioning.
Aggregate type	10 mm NMAS aggregates; limestone: predominant mineral co

Aggregate type	10 mm NMAS aggregates; limestone: predominant mineral composition was calcite (97%); granite: predominant mineral
	compositions (90%) including quartz (33%), albite (28%), k-feldspar (17), and chlorite (12%)
Aggregate gradation, SS/PP	9.5/95
Mineral filler type	Granite and limestone
Bitumen	40/60 pen
Air void	4%, 6%, and 8%
Moisture conditioning	Moisture conditioning at 60 °C for up to 70 days followed by 70 days drying; dry storage at room temperature for 140 days.
	Moisture-conditioned specimens were removed from water bath for testing at following days: 8, 15, 30, 55, and 70 days
^a Number of replicates	Four 100 mm diameter by 60 mm thick gyratory compacted specimens

NMAS = nominal maximum aggregate size; k-feldspar = potassium dominated feldspar; SS = sieve size in mm; PP = percent passing sieving size. ^a For each aggregate, mineral filler and conditioning type.



Fig. 1. Test set-up used for asphalt mixture moisture conditioning. Asphalt mixture test specimens sitting in deionized water at 60 °C for up to 70 days.

Table 2

Physico-chemical properties of the asphalt mixture constituents including surface free energy and thermodynamic work of adhesion and debonding of aggregate-bitumen bonds.

Material	SFE components (mJ/m ²)					$TWA_{AB} (mJ/m^2)$	$TWD_{AWB} (mJ/m^2)$
	γ^{LW}	γ^+	γ^{-}	γ^{AB}	γ^{T}		
Bitumen	30.6	0	2.4	0	30.6	N/A	N/A
Granite A	67.8	164.0	123	284.1	352.5	131	-109
Limestone A	75.2	109.0	49.9	148.5	222.7	128	-51

TWA_{AB} = thermodynamic work of adhesion between aggregate and bitumen; TWD_{AWB} = thermodynamic work of debonding aggregate—bitumen bond in the presence of water; N/A = not applicable. γ^{LW} = Lifshitz-van der Waals; γ^* = dispersion component; γ^- = polar component; γ^T = total surface free energy

2.4. Indirect tensile strength testing of asphalt mixtures

Indirect tensile strength (ITS) tests were performed in accordance with EN 12697-23 [19] on the 100 mm diameter by 60 mm thick specimens under the following conditions. Multiple sets of mixtures that have been subjected to different conditioning regimes (wet and dried or dry) were tested. They included (1) specimens stored in the dry state at room temperature for 116 days to represent the unconditioned state and (2) specimens that were dried for 47 days following the initial 70-day moisture conditioning. Comparison of the tensile strength data of the mixtures for the different conditioning states yielded additional evidence that supported the reversibility of moisture-induced damage in asphalt mixtures. The indirect tensile strength tests were conducted at 20 °C using a loading rate of 50 mm/min. The indirect tensile strength (σ) of the mixtures were calculated using Eq. (2) where *P* is the peak load, *D* the diameter of the specimen, and *t* the thickness of the specimen.

$$\sigma = \frac{2P}{\pi Dt} \tag{2}$$

3. Results and discussion

3.1. Indirect tensile stiffness of asphalt mixtures

Table 3 list the stiffness of the asphalt mixtures after exposure to water at 60 °C for durations ranging from zero to about 70 days. Each data point represents the average of four replicate specimens. Also shown in Table 3, for each data point, is the standard deviation. Several observations about the effects of moisture on stiffness reduction of asphalt mixtures can be deduced from Table 3. First, moisture conditioning resulted in significant reduction in mixture stiffness with respect to the initial stiffness. For example, the average stiffness of the LA + LF mixtures with 4.5% air voids, changed from 7257 MPa to 2111 MPa after 69 days of moisture conditioning, a reduction of about 71%. Similar results were obtained for mixtures at the different air void contents. Mixtures with the highest air voids suffered higher reduction in strength compared with mixtures with lower air voids. For examples, the stiffness of LA + GF with 3.8% air voids retained about 71% of stiffness for LA + GF with 7.9% air voids, after 55 days of moisture conditioning.

The effect of moisture conditioning on stiffness reduction was more severe in the mixtures containing granite mineral filler than mixtures that contained limestone filler, especially at the highest air void levels (Table 3). Overall, depending on mineral filler type and air void level, stiffness modulus of asphalt mixtures exposed to 55 days of moisture conditioning at 60 °C reduced by 11–71%. The combined effects of moisture and air void levels on stiffness degradation was not linear, mixtures with 6% nominal air voids performed better (higher retained stiffness) than mixtures with either 4% or 8% air voids. For example, considering the mixtures

Table 3 Stiffness, MPa, a	t 20 °C for asphalt mix	tures exposed to	o water at 60 °C. Each da	ata point represents the	average results of four	specimens plus/minus	one standard deviation.
Mix ID	Air voids (%)	Conditioni	ng time (days)				

IVIIX ID	All Volus (%)	conditioning time (days)					
		0	8	15	30	55	69
LA + LF	4.5	7257 ± 1165	4769 ± 399	5028 ± 670	5949 ± 1243	4820 ± 561	2111 ± 702
	6.6	6156 ± 1959	4769 ± 671	4543 ± 625	5048 ± 1255	5471 ± 902	2799 ± 736
	8.3	6244 ± 1715	5022 ± 910	6107 ± 1374	3877 ± 1691	3961 ± 821	2532 ± 977
LA + GF	3.8	7006 ± 1667	6434 ± 1435	6434 ± 1504	6033 ± 1425	4944 ± 1362	2573 ± 1267
	6.5	5372 ± 1353	4762 ± 1156	5298 ± 1711	4608 ± 579	3767 ± 887	2552 ± 412
	7.9	5024 ± 743	3834 ± 502	3850 ± 900	4048 ± 1121	1926 ± 555	2255 ± 995

at 6% air voids, which is the typical air voids found in newly constructed pavements, the shapes of the stiffness degradation curves appear to be nonlinear and sigmoidal with three stages that can be described as primary, secondary, and tertiary (Fig. 2). Subtle difference in performance could be seen in Fig. 2 where the duration of the secondary stage appears longer in LA + LF compared to LA + GF. As discussed later, the shape of the plots in Fig. 2 could be used to characterize moisture-induced stiffness degradation in asphalt mixtures.

The longer the time it takes a mixture to reach a given retained stiffness level, the better the resistance of the mixture to moisture induced damage. Thus the rate of stiffness degradation as a function of conditioning could be used to characterize the sensitivity of asphalt mixtures to moisture exposure if a suitable model could be fitted to the stiffness data. Eq. (3), a composite exponential and sigmoidal function is one such model, where *S* is the stiffness after



Fig. 2. Effects of moisture conditioning and air void levels on retained stiffness for asphalt mixture tested at 20 °C. The unique sigmoidal shape of the stiffness degradation curves could be used to characterize mixture sensitivity to moisture. (a) LA + LF. (b) LA + GF.

a given level of moisture exposure, k_i (i = 1-3) the model parameters, S_0 the initial stiffness, and θ the conditioning time in days.

$$S = \frac{k_1 S_0 e^{k_2 \theta}}{k_1 + S_0 (e^{k_2 \theta} - 1)} - k_3 \theta^4$$
(3)

The model in Eq. (3) was fitted to the experimentally determined stiffness degradation data presented in Fig. 2. Table 4 lists the fitted model parameters obtained. The correlation between the predicted stiffness degradation and the measured stiffness degradation was generally good as shown in Table 4. The differences in the model parameters did not vary significantly with respect to mineral filler type. However, for each mixture type air void content appears to influence stiffness degradation irrespective of filler type; mixtures compacted to 6.0% air voids appear to be more resistant to stiffness degradation from moisture exposure than at either the 4% or 8% air void level (lower k_3 values).

The plots shown in Fig. 3 (and also the original data shown in Fig. 2) can be used to explain the effects of moisture on stiffness degradation of asphalt mixtures. The sigmoidal nature of the plots in Fig. 3 suggests the degradation process may involve multiple stages (with different degradation rates) and is typical of stiffness degradation due to plasticization of an organic adhesive by moisture [20]. The first stage is associated with short conditioning time where the damage effect of moisture is a rapid loss of stiffness. During the second stage, the rate of stiffness degradation slows or even increases slightly in some mixtures. The duration of the secondary stage was sensitive to both mixture type and air void levels. This is clearly seen in Fig. 2 for LA + LF with 6% air voids where the second stage is significantly extended compared with mixtures with 4% and 8% voids. During the third stage, there is rapid loss of stiffness with moisture conditioning. The nature of the plots also suggests the damage is mainly cohesive with little adhesive damage. As discussed earlier (Table 2) when a material like bitumen with a low surface free energy is bonded to a high surface free energy substrate like aggregate, the thermodynamic work of adhesion is greater than the cohesion within the bitumen. Thus during the initial moisture conditioning (equivalent to low moisture content) of an asphalt mixture, it is expected that the weak link will be cohesion in the bitumen (or more appropriately the mastic) and not the bitumen-aggregate interface. If the latter assertion is true, i.e. cohesive damage dominate the stiffness degradation, then we should expect some stiffness lost due to moisture conditioning to be recovered upon specimen drying as discussed next.

3.2. Reversible stiffness degradation of asphalt mixtures

The sigmoidal nature of the plot of stiffness degradation with conditioning time (Figs. 2 and 3) suggests a plasticization process. As noted earlier, stiffness degradation resulting from the plasticization of some organic materials by moisture can be reversed if a previously wet specimen is subsequently dried. Fig. 4 shows that this

Predicted model parameters for stiffness degradation in asphalt mixtures exposed to water at 60 °C for 70 days.	s.

Mix ID	Air voids (%)	Model parameters			
		k_1	<i>k</i> ₂	k ₃	
LA + LF	4.5	5405.2993	3.99999	0.000133	0.90
	6.6	5037.1764	3.99999	0.000078	0.71
	8.3	5037.7981	3.99999	0.000113	0.78
LA + GF	3.8	6369.3637	3.99999	0.000166	1.00
	6.5	4891.6391	3.99999	0.000106	0.96
	7.9	3722.8922	3.99999	0.000083	0.79



Fig. 3. Sample predicted stiffness degradation of asphalt mixtures using exponential sigmoidal model in Eq. (2). Data were from mixtures containing 4% nominal air voids. The shapes of the plots are typical of cohesively dominated stiffness degradation resulting from a plasticization process.

was indeed the case for the asphalt mixtures tested in this study. As shown in Fig. 4a, stiffness of specimens with no moisture exposure (dry) decreased by about 64% from an average of about 6552 MPa to about 2481 MPa after 69 days of moisture conditioning (wet). When the Wet specimens were subsequently dried for 47 days under ambient conditions of 20 °C and 50% RH (Dryback), almost all the lost stiffness was recovered as the mixture stiffness rose to 6561 MPa. Similar trends in the reversibility of moisture-induced stiffness degradation can be seen in Fig. 4b. The results shown in Fig. 4 is significant as it demonstrates the existence of reversible stiffness degradation in asphalt mixtures. The results are also significant as they might help explain the discrepancy that exists between some laboratory test results that suggest poor moisture sensitivity and actual field performance in which the same mixture performs well.

It is interesting to note the similarity between the stiffness degradation obtained from physical testing (moisture conditioning and mechanical testing) of 64% and the predicted strength reduction obtained from applying changes in relative permittivity based on the chemical bonding theory (63%) as previously discussed (Eq. (1)).

3.3. Indirect tensile strength of asphalt mixtures

Stiffness data presented in Figs. 3 and 4 suggest plasticization might be a key factor in moisture-induced stiffness degradation in asphalt mixtures. The objective of performing the tensile strength tests was to determine if reversibility of moisture-induced degradation observed for stiffness could also be seen with respect to tensile strength of asphalt mixtures. To achieve this objective, two sets of mixtures were considered. The first set of specimens were the same as those labeled Dryback in Fig. 4, i.e. these mixtures were conditioned for 70 days in hot (60 °C) water and



Fig. 4. Stiffness of asphalt mixtures (LA + LF and LF + GF) subjected to three conditioning regimes showing reversible moisture-induced stiffness degradation. Dry = original specimens without moisture exposure; Wet = specimens subjected 69 days of moisture conditioning at 60 °C. Stiffness was determined at 20 °C. Each mixture type (LA + LF or LF + GF) was compacted to three different nominal air voids 4%, 6%, and 8%. (a) LA + LF. (b) LA + GF.

then subsequently dried for another 47 days. The second set of specimens were stored dry at room temperature for about 116 days (Original). All the tensile strength tests were conducted at 20 °C using a loading rate of 50 mm/min. During each tensile strength test, applied load (stress) and the corresponding cross-head deformation (strain) were monitored continuously. The peak stress and the strain at the peak stress were used to define failure in the mixtures. Fig. 5a and b shows the results of the tensile strength for Dryback and Original specimens, respectively. A clear trend of decreasing tensile strength with increasing air void content can be seen. For the Dryback mixtures (Fig. 5a), tensile strength was slightly higher in LA + GF than in LA + LF, which is similar to the results obtained for the stiffness tests.

The differences in tensile strength observed in the Original mixtures for LA + LF and LF + GF were not significantly different,



Fig. 5. (a) Indirect tensile strength of asphalt mixtures conditioned in water at 60 °C for 69 day and then dried for 47 days (=116 days of conditioning). (b) Indirect tensile strength of asphalt mixtures conditioned in at ambient conditions (20 °C and 50% RH) for 116 days. Tensile strength was obtained using a loading rate of 20 mm/ min and a testing temperature of 20 °C.

however, a decreasing trend of tensile strength with increasing air voids is similar to the Dryback mixtures. Therefore, to compare the two set of mixtures, the tensile strength values were normalized with respect to air voids. The results are shown in Fig. 6 where the normalized indirect tensile strength (normalized by percent air void content) is plotted against conditioning regime (Dryback or Original). The error bars represent plus or minus one standard deviation. For obvious reasons, the variability in tensile strength for the Dryback mixtures are higher than the Original mixtures. The results demonstrate that the differences in tensile strength for the two cases are not statistically significant. It can be concluded that tensile strength of asphalt mixtures subjected to certain moisture conditioning regimes are recoverable upon mixture drying. The results provide additional evidence in support of recoverability of moisture induced damage in asphalt mixtures. The significance of these findings is that reversible moisture-induced damage which is an important factor to consider during the analysis and design of asphalt pavements for durability can be quantified experimentally.

3.4. Strain at failure

A key manifestation of moisture sensitivity in asphalt mixtures is the reduction in tensile strength and the accompanying increase in strain at failure with moisture conditioning. Assuming the damage induced in the asphalt mixtures conditioned in water at 60 °C was reversible, then it follows that the strain at failure upon



Fig. 6. Recoverable tensile strength in moisture-conditioned asphalt mixtures. Comparison of indirect tensile strength (ITS) of asphalt mixtures subjected to two conditioning regimes: Dryback and Original. Dryback mixtures were moisture conditioned at $60 \,^{\circ}$ C for 69 days followed by 47 days of drying under ambient conditions. Original mixtures were stored at room temperature for 116 days. ITS was normalized by percent air voids of the mixtures. Error bars represent plus or minus one standard deviation.

Table 5

Recoverable tensile strain at peak load in mixtures previously moisture condition compared with un-conditioned asphalt mixtures.

Condition	Mix ID	Air voids (%)	Strain at peak load	
			Mean	Standard deviation
Original	LA + LF	5.6	0.0439	0.0017
-		6.8	0.0359	0.0034
		9.0	0.0316	0.0024
	LA + GF	5.3	0.0273	0.0023
		7.2	0.0356	0.0030
		7.5	0.0429	0.0112
Dryback	LA + LF	4.5	0.0260	0.0070
		6.6	0.0345	0.0066
		8.3	0.0356	0.0022
	LA + GF	3.8	0.0340	0.0027
		6.5	0.0310	0.0042
		7.9	0.0309	0.0058

Dryback = mixtures that were moisture conditioned at 60 $^{\circ}$ C for 69 days followed by 47 days of drying under ambient conditions. Original = mixtures that were stored at room temperature for 116 days.

drying (Dryback) should be similar to the unconditioned mixtures (original). As shown in Table 5, for the asphalt mixtures tested, this was indeed the case as the average strain at peak strength for the Dryback mixtures (0.0320) were comparable to that of the Original mixtures (0.0362). The results provide further evidence yet of the existence of reversible moisture-induced degradation in asphalt mixtures.

3.5. Moisture diffusion

The previous discussion demonstrated that stiffness degradation in asphalt mixtures that have been exposed to moisture can be reversed if the mixture is re-dried. This led to the conclusion that for the asphalt mixtures considered, stiffness degradation occurred in the mastic as cohesive damage as opposed to adhesive damage where water attacks the interface. To determine the possibility of water diffusing through the mastic into the interface, two key parameters are needed: (1) diffusion coefficient of the mastics, and (2) thickness of the mastic films. Moisture diffusion coefficient of asphalt mastics at 20 °C have been determined in previous studies [21,22].



Fig. 7. Micro CT scans of dense graded asphalt mixture. (a) Original image, brighter areas represents aggregates. (b) Segmented image showing mastic moisture diffusion paths (white areas) around the aggregate (darker areas) particles. Diameter of specimen approximately 100 mm diameter.

To determine a reasonable estimate of mastic thickness and length of diffusion path, samples of the asphalt mixtures internal images were obtained using a micro-CT scanner. A VENLO H-350/ 225 scanner (X-Tek Industries, Tring, Hertfordshire, U.K.) with two micro-focal X-ray sources (225 and 350 kV), 1829 IA X-ray intensity (640 W power), and 1-mm collimation (slice thickness) was used to scan the asphalt mastic samples at a resolution of 0.083 mm/pixel. The obtained images were processed using image processing tools to segment the image into its three key components: air, mastic, and aggregates. Fig. 7a shows the CT image before image processing; here the brighter, the image, the more dense is the material. Therefore, the outline of the denser aggregates can be clearly seen. The darkest blobs are air voids. The segmented image is shown in Fig. 7b. Here, the whiter region corresponds to mastic and air voids, while the darker regions represent the aggregates. Careful comparison of Fig. 7a with b enabled the identification of the mastic films. Following the identification of the mastic films an estimate of their thickness ranges were made as 0.633-1.899 mm.

To estimate the amount of time required for a water molecule to reach an interface the average diffusion path was assumed. The diffusion path (i.e. the path a water molecule entering the mix from the outside will traverse to reach an aggregate interface) was assumed to be 25 mm. From literature, diffusion coefficient of asphalt mastics has been reported in the range of $0.3-1.5 \times 10^{-12}$ m²/s so a value of 1.5×10^{-12} m²/s was chosen. Using Fick's law [23], for the conditions specified, it would take about 473 days for water to reach the interface of an aggregate located 25 mm from the edge of a specimen. Thus it is reasonable to assume that for the conditions used in this study, degradation of the aggregate–mastic interface is minimal. The results supports the assertion of plasticization of the aggregate–bitumen interface may be responsible for the observed phenomenon of reversible degradation.

4. Conclusions and recommendations

The durability of asphalt mixtures immersed in water at 60 °C was investigated to better understand the moisture damage phenomenon by using measurements of indirect tensile stiffness as a function of exposure time, mineral filler, and percent air voids. The following conclusions were reached based on the results presented in this paper:

 Stiffness generally decreased with conditioning time (and consequently water content) except at low exposure times where low water content caused slight increases in stiffness. Depending on mineral filler type and air void levels, stiffness modulus of asphalt mixtures exposed to 55 days of moisture conditioning at 60 °C reduced by 11–62%.

- After 70 days of moisture conditioning at 60 °C, asphalt mixtures containing granite filler were less durable than similar mixtures containing limestone filler suggesting the influence of mineralogical and physico-chemical on durability asphalt mixture exposed to moisture.
- Durability was negatively affected in asphalt mixtures with high air voids irrespective of mineral filler type which suggest an association between durability and amount of water exposure.
- The effect of moisture exposure on durability was found to be reversible; moisture conditioned asphalt mixtures that had lost up to 80% of the initial stiffness upon drying fully recovered both their stiffness and tensile strength at 20 °C.
- The reversible moisture-induced stiffness degradation indicates a plasticization process. It also suggests cohesive rather than adhesive failure dominates the durability of asphalt mixtures under long-term moisture exposure.
- Estimates of mastic thickness and length of diffusion paths obtained from image analysis of micro-CT scans of the asphalt mixtures internal structure suggest, for the duration of moisture conditioning used, moisture diffusion was mainly restricted to the bulk mastic and not the aggregate-mastic interface.
- The slight but repeatable increase in stiffness at low water content may be an indication of stress relaxation.

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