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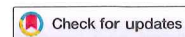
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Influence of bitumen and aggregate polarity on interfacial adhesion

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The quality of the bitumen–aggregate interface formed in an asphalt mixture is dictated by the physicochemical properties of both materials, such as surface polarity. In order to better understand the role of polarity on interfacial adhesion between bitumen and aggregate, a systematic study was conducted by designing bitumens with different polarities. These bitumens were produced by the addition of diatoms and hydrophobic diatoms in varying concentrations. It was found that by manipulating the polarity of bitumen it is possible to design materials that result in bitumen–aggregate pairs that are more compatible with each other and resistant to debonding. On the other hand, basicity of aggregates is also highly related to obtaining asphalt mixtures with improved durability. The findings from this paper help advance the fundamental understanding of the adhesive behaviour between bitumens and aggregates. More importantly, these findings can be used to design optimal anti-strip agents for using in different asphalt mixtures.

Keywords: water-resistant interfaces; durability; asphalt mixture; surface energy

Introduction

One of the factors that affect the durability of asphalt mixtures is the resistance of the bitumen and bitumen–aggregate interface to moisture damage. Moisture damage is recognised as one of the most severe distresses on pavements (Caro, Masad, Bhasin, & Little, 2010). This is of special interest in regions with high levels of ambient moisture or rainfall in specific seasons or throughout the year. Several mechanisms drive moisture transport within the asphalt mixture and ultimately to the bitumen–aggregate interface. Once the moisture arrives at the bitumen–aggregate interface a detachment of the bitumen from the aggregate surface occurs and hence, the weakening of the mastic (Caro, Masad, Bhasin, & Little, 2008). The magnitude of moisture damage that can affect the pavement depends in part on the physical and chemical properties of its components (Gorkem & Sengoz, 2009). Several research efforts have focused on modifying the bitumen with materials that can improve its adhesion with aggregate (Boulangé, Bonin, & Saubot, 2013). However, in order to design the optimal modifier for any given bitumen – aggregate pair, it is necessary to recognise that adhesion is a complicated process. Existing studies suggest that durable adhesion is predominantly achieved by the polar components in bitumens that tend to interact strongly with the aggregate surface (Curtis, 1992; Robertson, 2000). One of

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the goals of this study was to quantitatively estimate the relative contributions of the non-polar (dispersive) and polar components of bitumens and aggregates to the quality of their interfacial bond. In order to achieve this goal, it is important to measure the polarity of materials involved. A simple way to obtain this information is estimating surface free energy (SFE) of bitumens and aggregates, which in turn provides an indication of their polar and non-polar surface characteristics.

To evaluate the contribution of polar interactions on adhesion and debonding processes, it was necessary to incorporate bitumens and aggregates with different polarities. In the case of aggregates, this was achieved by choosing materials from different sources and compositions. In the case of bitumens, increasing polarity can be achieved by modification, using inorganic materials such as diatoms, a type of algae with a cell wall composed mainly of silicon dioxide. The use of inorganic materials is promising not only because of their chemical nature, but because of the good performance shown in previous research (Cong, Chen, & Chen, 2012; Yao et al., 2013; Yi-qiu, Lei, & Xing-you, 2012). In addition to diatoms, a second material was also included: hydrophobic diatoms. Hydrophobic diatoms are regular diatoms with a hydrocarbon-functionalised surface. The latter material is a hybrid, which means that it has both polar and non-polar characteristics, and is therefore expected to increase the polarity of bitumens, as well.

The first step to achieve the objective of this study was to create a set of bitumens with different levels of polarity by addition of diatoms and hydrophobic diatoms. To the best of the authors' knowledge, a unique feature of this study is the use of these systematically engineered bitumens with varying levels of polarity. The second step was to measure the surface characteristics of these bitumens and aggregates and use them as a basis to evaluate the relationship between polarity and the quality of the interfacial bond in wet and dry conditions.

Background

Physicochemical properties of materials: SFE

The SFE is an intrinsic property of all solids and liquids. It is defined as the energy required to create a new surface of unit area in vacuum. The knowledge of this property allows understanding the formation of interfaces, such as the one formed between the bitumen and aggregate in an asphalt mixture.

The theory developed by van Oss, Chaudhury and Good was applied to quantitatively decompose the SFE of materials into its polar and non-polar components. This theory states that the total SFE (γ) is the sum of two chemical characteristics of materials: the dispersive (non-polar) character (γ^{LW}) and the polar character (γ^{AB}). The latter in turn is divided into two parts: the acidic (γ^+) and the basic (γ^-) components, as shown in Equation 1 (van Oss, Chaudhury, & Good, 1988).

$$\gamma = \gamma^{LW} + \gamma^{AB} = \gamma^{LW} + 2\sqrt{\gamma^+\gamma^-}. \quad (1)$$

Once the bitumen coats the aggregate surface during mixing, a new interface is formed. The correct characteristics of this interface depend on the interactions taking place between both materials. It is possible to estimate the strength of these interactions by means of the work of adhesion (W_{AB}) (Equation 2).

$$W_{AB} = \gamma_A + \gamma_B - \gamma_{AB}, \quad (2)$$

where γ_A and γ_B refer to SFE of the aggregate and bitumen, respectively, and γ_{AB} refers to the new aggregate-bitumen interface.

Unfortunately, the bitumen-aggregate interface is easily breached by water. Then, it is thermodynamically favourable for water to displace and completely detach the bitumen. The energy

released or work done by the system in this process is known as work of debonding (W_{ABW}^{wet}) (Equation 3).

$$W_{ABW}^{wet} = \gamma_{AW} + \gamma_{BW} - \gamma_{AB}, \quad (3)$$

where γ_{AW} and γ_{BW} refer to the new interfaces formed between aggregate and water and bitumen and water, respectively.

Moisture-induced damage occurs due to a complex interaction of adhesion and detachment processes. In order to estimate the potential of moisture damage for a given bitumen–aggregate pair, Little and Bhasin (2006) proposed a parameter denoted as Energy Ratio (ER) which relates the work of bonding and debonding into a single indicator (Equation 4)

$$ER = \frac{W_{AB}}{W_{AB}^{wet}} \quad (4)$$

The previous knowledge of Energy Ratios allows predicting the moisture damage resistance for any given bitumen–aggregate combination: the higher the ER, the higher the durability (Bhasin, Masad, Little, & Lytton, 2006). In this study W_{AB} , W_{ABW}^{wet} and ER have been chosen to evaluate the effect of the additives proposed to improve the moisture damage resistance of the bitumens.

Chemical nature of materials and interfaces

The formation of a new bitumen–aggregate interface is mainly a physicochemical process. Therefore, knowledge on chemical properties of the materials involved, such as polarity, is required to understand the feasibility with which the formation of the interface will take place. In other words, the quality of coating of one material by another depends on the compatibility of their surface chemistries.

It was hypothesised that by manipulating the polarity of bitumens it is possible to produce bitumens with an improved compatibility to aggregates. This was achieved by using materials with different polarities as additives in asphalts. The resulting bitumens were then evaluated for their adhesion and debonding characteristics by applying the SFE theory discussed previously.

Note that the bitumen is only half of the interface formed. It is necessary to consider the chemical characteristics of the aggregates, as well. By doing so it is possible to predict the tendency of some aggregates to prefer some bitumens over others and the reason for such behaviour.

Materials and methods

Synthesis of hydrophobic diatoms

Surface functionalisation of diatoms was performed based on previous work (Fatoni, Koesnarpardi, & Hidayati, 2010). Briefly, 35 g of diatoms were suspended into 150 mL of toluene on a 500 mL three-neck round-bottom flask. Next, 10 mL of triethoxyoctylsilane was added with a syringe and the suspension was stirred and refluxed overnight. The solid obtained was filtered and washed with 50 mL portions of toluene, ethyl ether and ethanol and allowed to stand on a vacuum desiccator at 60°C for 6 hours.

Characterisation of additives

Infrared spectra

In order to confirm the desired functionalisation of diatoms, the FTIR measurements of the hydrophobic diatoms were performed and compared to the one of un-functionalised material.

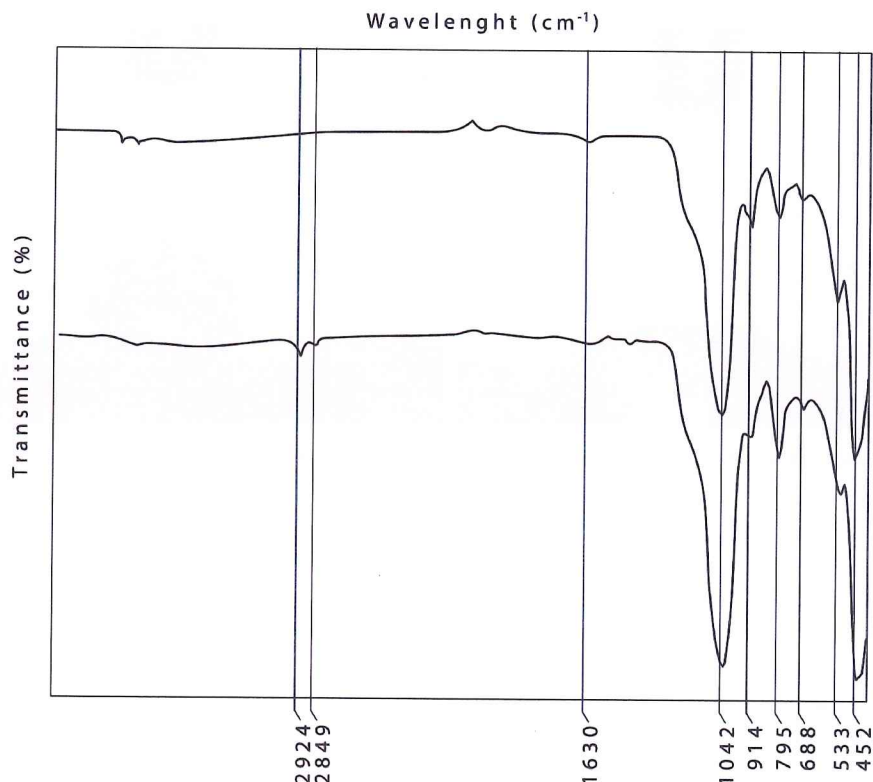


Figure 1. IR spectra for (a) diatoms and (b) hydrophobic diatoms.

The results are displayed in Figure 1. The bands located at 452; 795 and 1042 cm^{-1} correspond to the bending and stretching vibrations of O–Si–O groups, and the shoulder located on the 1042 cm^{-1} band is attributed to an out-of-phase stretching vibration of Si–O group (Jiang et al., 2014). The bands around 1600 cm^{-1} and 1630 cm^{-1} correspond to the stretching and bending vibrations of H–O–H, respectively (Cai et al., 2009). The bands at 914 and 938 cm^{-1} are related to the Si–O stretching vibrations, and the bands at 534 and 688 cm^{-1} are attributed to inorganic compounds (Jiang et al., 2014). Finally, the spectrum of hydrophobic diatoms shows the typical bands of C–H stretching vibration located around 2900 and 2800 cm^{-1} . These bands confirm the presence of the hydrocarbon chain introduced to the diatoms during the functionalisation.

Contact angle measurement

The hydrophobicity of functionalised diatoms was evaluated by means of contact angle measurements using a Ramè-Hart 250 goniometer and the Drop Image Advanced software. A disk of the hydrophobic diatoms was obtained and a drop of water was placed over the surface. The same procedure was carried out for the un-functionalised diatoms for comparison (Figure 2). The resulting contact angles were found to be on average 36° for un-functionalised and 135° for functionalised diatoms. These results show a significant change on the chemical nature of diatoms and demonstrate that after modification the diatoms become hydrophobic.

Thermal behaviour

To ensure that thermal decomposition of additives does not occur during bitumen modification, the thermogravimetric analysis of materials was performed using a TGA Q500. The obtained

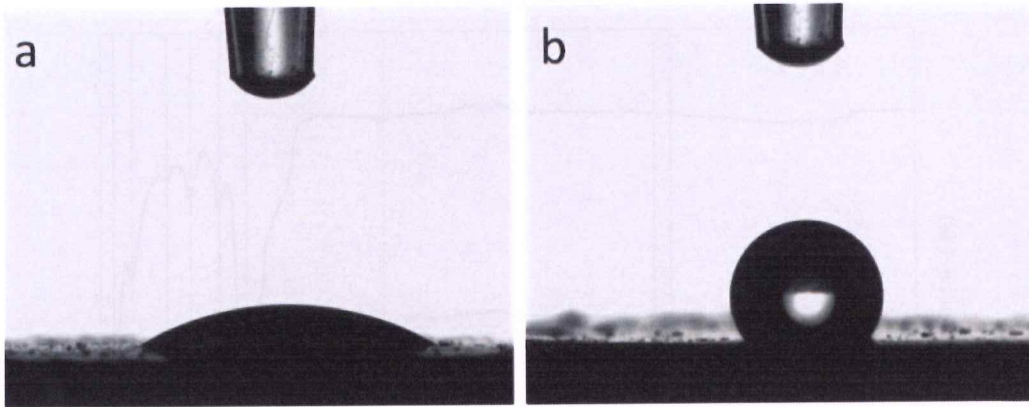


Figure 2. Drop of water over the surface of (a) diatoms and (b) hydrophobic diatoms.

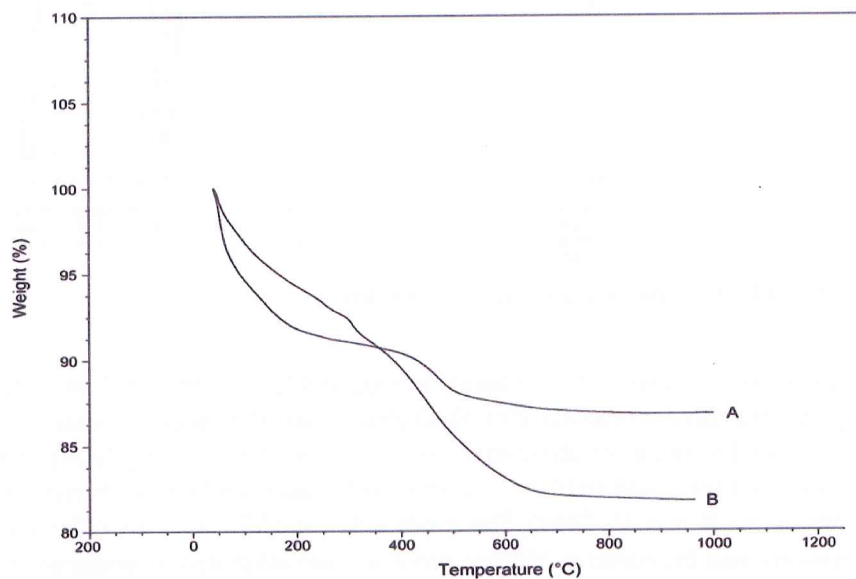


Figure 3. Thermograms for (a) diatoms and (b) hydrophobic diatoms.

thermograms are shown in Figure 3. For both materials, a loss of water around 100°C is observed and degradation of organic matter occurs between 200°C and 600°C. These results are in agreement with previous research (Jiang et al., 2014). From the hydrophobic diatoms' thermogram, it is observed that the weight loss of organic matter is higher compared to the diatoms alone. This is consistent with the fact that the hydrophobic diatoms carry a higher amount of organic matter due to surface functionalisation. The residue corresponds to inorganic materials such as silica and other oxides.

Characterisation of asphalts and aggregates

The neat bitumen was modified with diatoms and hydrophobic diatoms, separately, in concentrations of 2%, 4%, 6% and 10% by weight of bitumen. The modification was carried out at 175°C and the additives were incorporated with a low shear mixer during 1.5 hours.

The surface energy components of bitumens and aggregates were determined by means of contact angle measurements (Wei & Zhang, 2012). A full explanation of the mathematical

Table 1. Bitumens and aggregates characterisation.

Material	Identifier	Surface free energy				
		γ^{TOTAL}	γ^{LW}	γ^{AB}	γ^+	γ^-
Bitumens	Neat	20.1	19.0	1.03	0.128	2.07
	2% D ^a	17.5	16.7	0.836	0.0380	4.59
	4% D	15.0	11.8	3.15	0.521	4.76
	6% D	13.2	6.94	6.26	2.93	3.35
	10% D	14.0	10.2	3.81	1.31	2.78
	2% HD ^b	15.9	12.9	3.02	0.790	2.88
	4% HD	13.3	8.63	4.69	2.12	2.59
	6% HD	15.4	11.0	4.42	1.27	3.84
	10% HD	17.6	15.7	1.95	0.536	1.82
	Aggregates	L-1 ^c	42.1	39.4	2.77	0.226
L-2		54.7	46.9	27.9	0.572	26.5
G-1 ^d		51.2	23.3	3.77	5.87	33.2
G-2		44.1	40.3	7.79	0.0710	50.2

^aD = diatoms-modified bitumens.

^bHD = hydrophobic diatoms-modified bitumens.

^cL = limestone.

^dG = Gravel.

Table 2. Approximated chemical composition of aggregates

Aggregates	Minerals contained
L-1	Calcite (Mg, Ca)CO ₃
L-2	Calcite (Mg, Ca)CO ₃
G-1	Albite, Na(AlSi ₃ O ₈); Anorthite, (Ca,Na)[(Al,Si) ₄ O ₈]; Hauyne, (Ca,K,Na) ₈ [(Al,Si) ₁₂ O ₂₄ (SO ₄) ₂]; Quartz, SiO ₂
G-2	Albitecalcian, (Ca _{0,5} ,Na _{0,5})[(Al _{1,5} Si _{2,5} O ₈)]; Anorthite, (Ca _{0,66} Na _{0,34})[(Al _{1,66} ,Si _{2,34})O ₈]; Sanidine, (K,Na)(AlSi ₃ O ₈)

transformations regarding the measurement of surface energy components can be found elsewhere (Aguiar-Moya et al., 2016; Arabani & Hamed, 2011; Hefer, Bhasin, & Little, 2006; Moghadas Nejad, Hamed, & Azarhoosh, 2013). Table 1 presents a summary of results from these measurements.

Finally, the chemical composition of aggregates was determined using X-ray powder diffraction with a Bruker D8 Advance instrument. The results are listed in Table 2.

Results and discussion

The introduction of an additive in the bitumen matrix has an immediate influence on its physico-chemical properties. This observation is evident from the surface energy data shown in Table 1. Generally, it is noticed from Table 1 that with the addition of the diatoms and hydrophobic diatoms to the neat bitumen, the dispersive component is diminished and the polar component is increased. This was not only expected, but designed. The use of diatoms as an additive will increase the polarity of bitumens, even for the hydrophobic diatoms.

Due to the changes in SFE, the interaction of bitumen with the aggregate in the coating process (W_{AB}), as well as the resistance of the bitumen–aggregate interface to debonding in the presence of water ($W_{\text{ABW}}^{\text{wet}}$) are expected to change.

First of all, it is necessary to recall that a higher value of W_{AB} implies a stronger adhesion during dry conditions between the two components (Little & Bhasin, 2006). Hence, it is desired to achieve bitumen–aggregate combinations that have a high magnitude of W_{AB} . For the proposed materials, the work of adhesion (W_{AB}) is presented in Figure 4.

In comparison with the neat bitumen, the additives seem to reduce the adhesion between bitumen and aggregates. In order to further understand this behaviour, the contribution of the polar and dispersive components for each bitumen–aggregate pair was computed. Figure 5 illustrates these results. As expected, for the control bitumen (and any given aggregate) the dispersive component was the most significant contributor to the work of adhesion. However, upon addition of the polar additives, the relative contribution of the polar components increased and the relative contribution of the dispersive component decreased. While this is expected, given the fact that the overall work of adhesion decreased (Figure 4), these results suggest that globally the non-polar or dispersive component plays an important role in the work of adhesion.

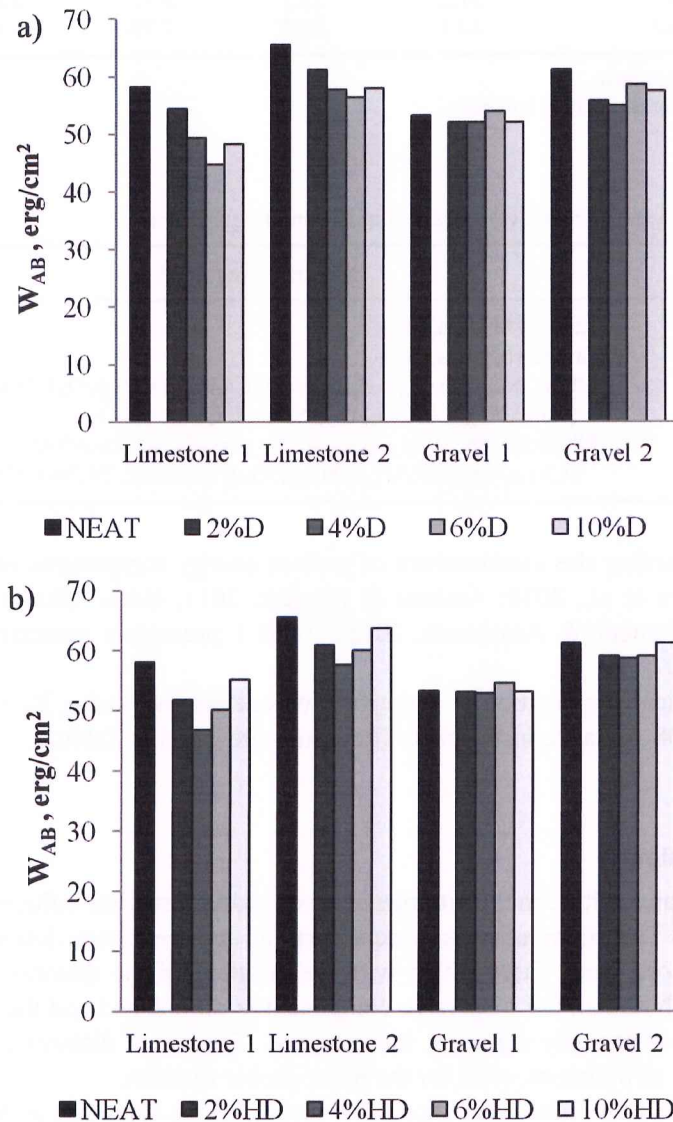


Figure 4. Work of adhesion (W_{AB}) for the pairs' aggregate and bitumens modified with (a) diatoms and (b) hydrophobic diatoms (Bars for control bitumens separated for clarity).

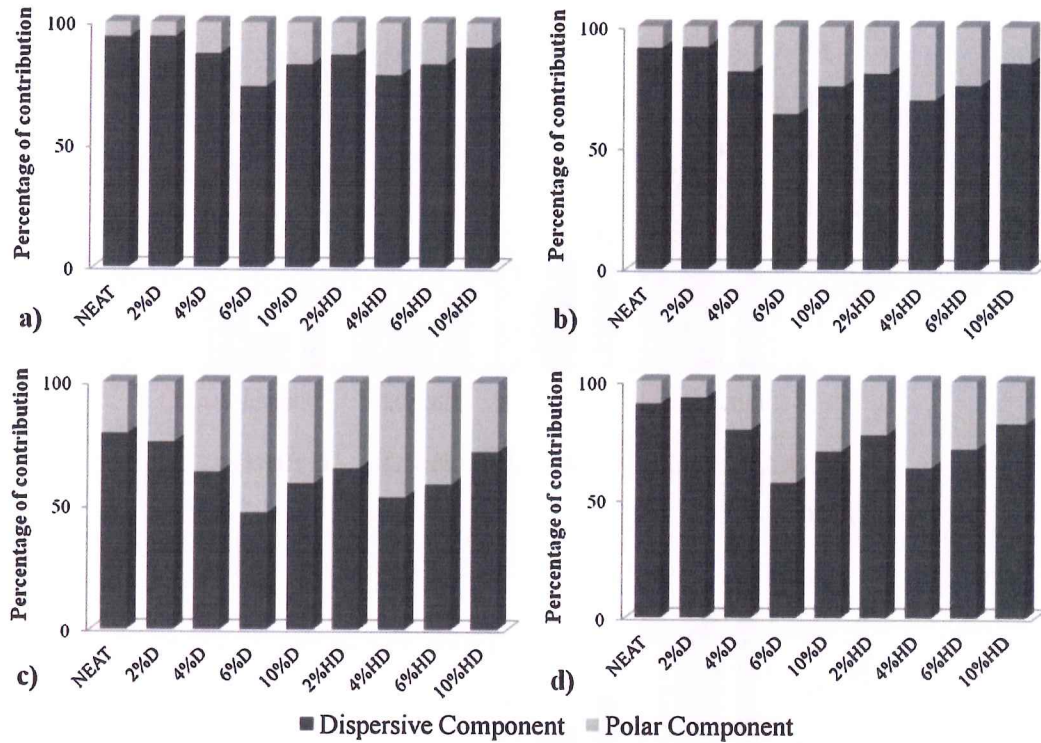


Figure 5. Contribution (in percentage) of dispersive and polar components of bitumens to adhesion when combined with (a) Limestone 1; (b) Limestone 2; (c) Gravel 1 and (d) Gravel 2.

The processes taking place on the adhesive interface formed between bitumen and aggregate are not fully understood. In fact, considering that bitumen is the adhesive that binds aggregates in a bitumen mixture, several adhesion theories have been proposed (Little and Jones, 2003). Among these theories, the fact that dispersive (van der Waals) forces dictate the bitumen–aggregate interaction is well recognised (Apegyei, Grenfell, & Airey, 2014; Baldi-Sevilla, Montero, Aguiar, & Loría, 2016) and is consistent with the discussion presented relative to Figure 5.

Another parameter that is relevant to evaluate the durability of the bitumen–aggregate interface is the work of debonding in the presence of water, W_{ABW}^{wet} . It is desired that W_{ABW}^{wet} be as low as possible so that the debonding process is unfavourable (Little & Bhasin, 2006). Figure 6 illustrates the W_{ABW}^{wet} for the proposed combinations.

It can be seen from Figure 6 that after modification the work of debonding decreases, which implies that the additives make the bitumen's detachment by water less favourable. It must be noted that additives were designed to increase the polarity of bitumens. In this sense, these results suggest that increasing polarity of bitumens is directly related to the reduction of debonding for the pairs proposed. This is essentially true if the neat bitumen is compared to the modified bitumens, individually. However, depending on the aggregate used, either the polar or dispersive components of the bitumen become important in the debonding process (Figure 7). Thus, a linear increase in polarity does not necessarily result in a linear reduction in resistance to debonding.

Finally, all of these changes in adhesion and debonding influence the resulting energy ratios (ER). Recall that the ER is the ratio between work of adhesion and work of debonding (Little & Bhasin, 2006). In other words, durability of a bond is dictated by the initial quality of the bond in dry condition, as well as by the resistance of this bond to separation induced by water. Most of the combinations involving modified bitumens produce higher ER compared to those of neat

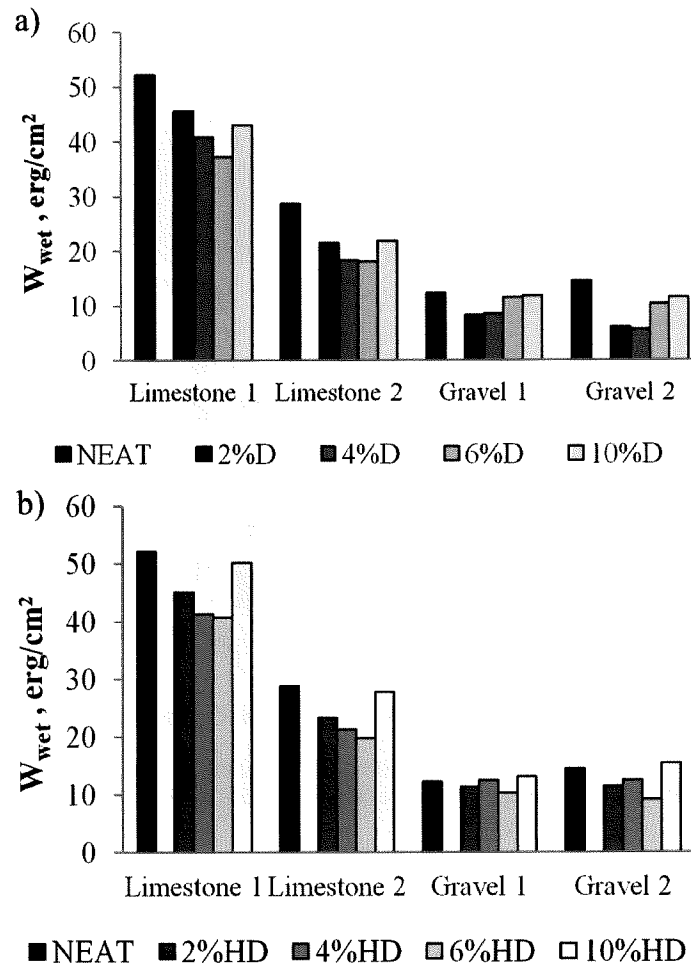


Figure 6. Work of debonding in the presence of water (W_{ABW}^{wet}) for the pairs' aggregate and bitumens modified with (a) diatoms and (b) hydrophobic diatoms.

bitumen (Figure 8). This means that the addition of diatoms or hydrophobic diatoms to a bitumen improves the durability of bitumen–aggregate pairs when any of the analysed aggregate sources proposed are used.

The most important observation from Figure 8 is that there is an optimum content of additive on bitumens to produce the highest ER. In the case of diatoms, this content is 4% for all of the aggregates, except for Gravel 1 (optimum content of 2%). The optimum content for hydrophobic diatoms is 6% for all aggregates. The results also show that a continuous increment in the additive does not necessarily result in a linear improvement of its properties.

These results also demonstrate that when developing chemical additives to enhance the adhesion characteristics of bitumen, it is important to consider the type and specific mineralogy of the aggregate. From Figures 4 and 7, it can be observed that W_{AB} and W_{ABW}^{wet} have different magnitudes based on the used aggregate source. The reason is that these values are highly related to the basic component (basicity) of the aggregates, as per Figure 9.

Although the correlation coefficient is low for W_{AB} , it is generally seen that aggregates with high basicity tend to produce interfaces with stronger adhesion and higher resistance against water. Although this observation seems to be interesting, it is not possible to generalise this finding using only four different aggregate types. For this reason, in order to further investigate this hypothesis, the SFE values of several aggregates from an existing database (Lytton,

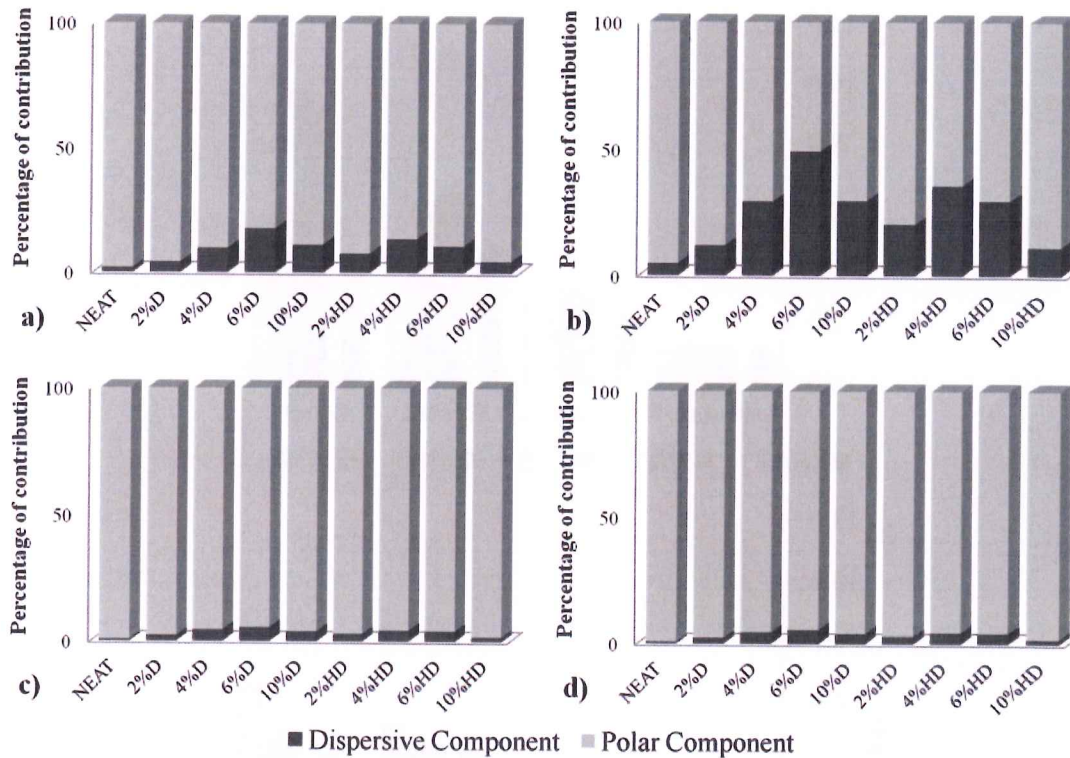


Figure 7. Contribution (in percentage) of dispersive and polar components of bitumens to work of debonding when combined with (a) Limestone 1; (b) Limestone 2; (c) Gravel 1 and (d) Gravel 2.

Masad, Zollinger, Bulut, & Little, 2005) were used in the same analysis as above (Figure 10). In such document, Lytton *et al.* described the production and evaluation of moisture damage susceptibility of several asphalt mixtures, and related the surface energy data to the field performance.

The trend followed by the combinations shown in Figure 10 supports the idea that the higher the basicity of aggregates, the higher the adhesion and water-resistance of the interfaces produced. In this sense, it is important to recall that the gravels produced the pairs with higher ER. Based on Table 2, these aggregates are chemically composed of mainly aluminium silicates, which provide to the aggregate its basic characteristics. On the other hand, the limestones are mainly composed of calcite, which is a basic mineral as well, but of lower basicity compared to the aluminium silicates of gravels. Therefore, the basicity of aggregates is proposed as a crucial characteristic that must be taken into account to design bitumen–aggregate combinations with improved durability. These results are in agreement with previous research that has identified that adding hydrated lime (a basic material) to acid aggregates significantly improves the resistance of the resulting mixture to moisture-induced damage (Airey, Collop, Zoorob, & Elliott, 2008).

Conclusions

The following are some key conclusions that can be drawn from this study:

1. Existing literature suggests that polar bitumens tend to have improved adhesion characteristics. However, results from this study demonstrate that typically additives that

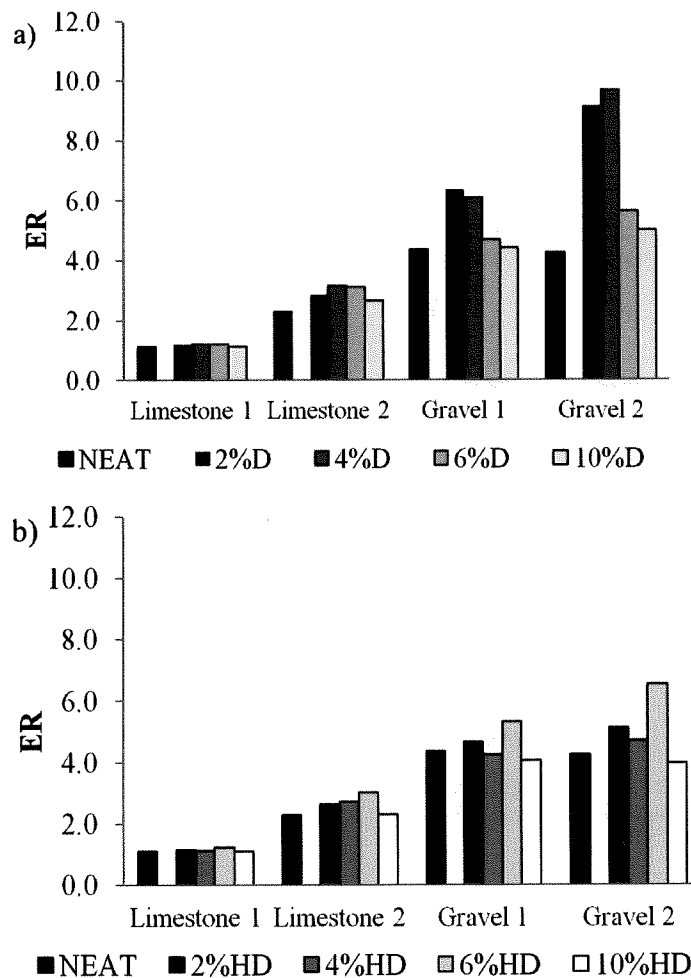


Figure 8. Energy ratios (ER) for the pairs' aggregate and bitumens modified with (a) diatoms and (b) hydrophobic diatoms

increase polarity of the bitumens tend to reduce the dry work of adhesion between the bitumen and the aggregate. However, additives that increase polarity of the bitumen also tend to reduce the ability of water to debond the bitumen from the aggregate surface. If the ER from previous studies is an indicator of resistance to moisture damage, then the polar additives must be designed such that the small loss in adhesion (under dry conditions) is more than compensated by the ability of the water to debond the bitumen from the aggregate surface.

2. The optimal level of polarity in a bitumen to improve its resistance to moisture damage is highly dependent on the type of aggregate selected. In other words, design of additives for bitumens cannot be done without considering the type of aggregate for which the additive is intended: there is no "one-fits-all" solution.
3. Basicity of aggregates appears to be an important factor in dictating the dry work of adhesion as well as the work of debonding in asphalt mixtures. Aggregates with higher basicity tend to interact strongly with bitumens and make the debonding process in wet condition less favourable. However, the polarity of a bitumen must be taken into account as well when choosing the aggregate source to produce asphalt mixtures with improved durability.

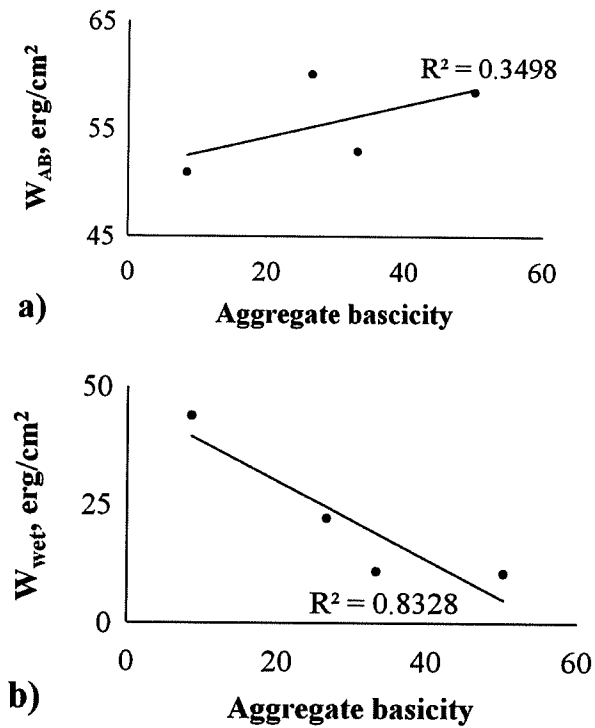


Figure 9. Effect of basicity of aggregate on (a) W_{AB} and (b) W_{ABW}^{wet} .

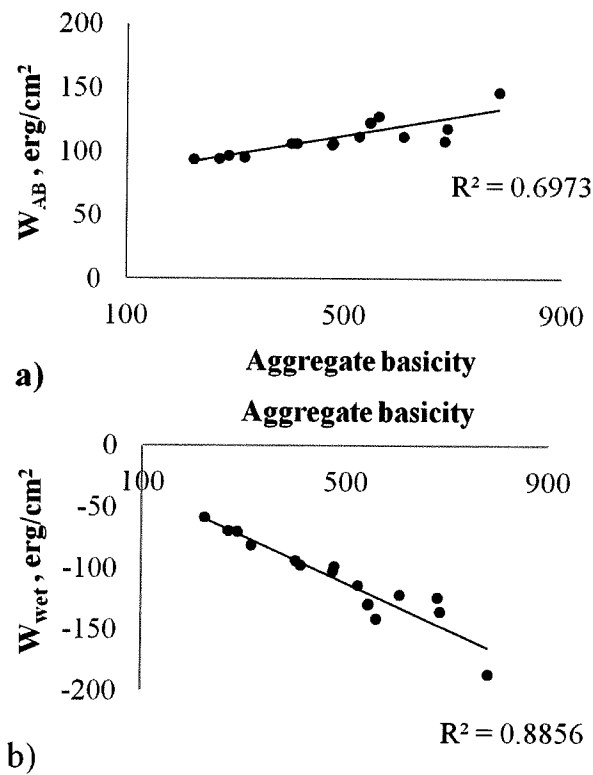


Figure 10. Effect of data base-aggregates basicity on the work of adhesion (W_{AB}). The values were calculated using the bitumens modified with diatoms and hydrophobic diatoms.

In summary, this study evaluated the durability of several different types of bitumen–aggregate interfaces. Although ER was used as the basis to quantify durability as a moisture damage indicator, further work including the use of mechanical tests on these bitumen–aggregate pairs to evaluate moisture resistivity is required.

Disclosure statement

No potential conflict of interest was reported by the authors.

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