



## Influence of Mixing Conditions of Modified Bitumen on Moisture Sensitivity of Asphalt Compounds

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### ABSTRACT

Modification of bitumen allows an improving the properties of bitumen, which can be a useful technique to enhance asphalt's characteristics. Therefore, this research intended to study the effect of mixing time of different percentages of epoxy resin, between 0 to 8%, doped into bitumens on the moisture sensitivity of asphalt compounds containing two types of aggregates, i.e. limestone and siliceous. For this aim, the Sessile Drop Method (SDM) was employed, and moisture resistant parameters associated to surface free energy concept such as surface free energy components, work of cohesion, work of adhesion, work of debonding, adhesion bitumen to aggregate, and wetting bitumen around aggregate surface were assessed. The results revealed that the effect of the mixing time of bitumen and epoxy resin on moisture resistance properties of asphalt compounds significantly depends on the percentage of epoxy resin used in bitumen mixture. For bitumens with 4 and 6% epoxy resin, adhesion and wetting parameters had a similar performance for both mixing times of 10 and 60 minutes. However, asphalt compounds containing 2 and 8% epoxy resin modified bitumens prepared for 10 min mixing time had a better adhesion and wetting performance than those prepared for the longer mixing time, 60 minutes. The effect of mixing time of bitumen modified with epoxy resin on the bitumen and asphalt compound' performance depend on the percent of additive in the bitumen.

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## 1. INTRODUCTION

The Moisture sensitivity of asphalt compounds is one of the most common damages that occurs in asphalt pavements [1–8]. The reduction in the pavement integrity due to moisture damage plays an important role in other types of distresses, such as rutting, fatigue cracking, and raveling [9].

There are three mechanisms in which moisture is capable of degrading the asphalt compound: (1) loss of cohesion within the bitumen, (2) adhesive failure between aggregate and bitumen, and (3) degradation of the aggregates. The cohesive failure occurs due to the rupture of bonds between molecules in the bitumen, and

the work of cohesion describes the resistance of bitumen against cohesive failure. On the other hand, adhesive failure happens when the bonds between bitumen and aggregate are ruptured, and work of adhesion explains the resistance of asphalt compounds against this type of failure. Moreover, the work of debonding is the measure of the work of adhesion between bitumen and aggregate in the presence of water [10].

The moisture damage is usually studied using experimental laboratory testing methods. A number of laboratory tests have been developed over the years to determine the moisture susceptibility of bituminous compounds. They include the modified Lottman test, Tunnickliff and Root method, Hamburg wheel tracking,

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asphalt pavement analyzer, saturation ageing tensile stiffness (SATS), Marshall immersion test, resilient modulus test, and immersion-compression test [11–14]. Nevertheless, these approaches are not able to examine fundamental features of materials to predict adhesion and wetting ability of each bitumen-aggregate composition [15]. An important material property concerning the moisture sensitivity of asphalt compounds is the surface free energy of the asphalt binder and the aggregate. The surface free energy of these materials can be used to determine work of cohesion, work of adhesion, and work of debonding [15]. In the surface free energy method, surface free energy properties of the materials are used to assess both the adhesion and wetting characteristics of bitumen and aggregate composition, and thereby the moisture sensitivity of asphalt compounds [15]. It was already shown that the surface free energy method is suitable for the selection of materials resistant to moisture [16–19]. There are different methods of measuring surface energy of asphalt compounds. In some researches, the surface free energy of bitumen and components of aggregates have been measured using the Wilhelmy plate method and the universal sorption device (USD), respectively [20–22]. On the same subject, the sessile drop method (SDM) has been used to measure the surface free energy of bitumen [23]. Using the SDM by a number of researchers, the effect of additives on the adhesion mechanism of bitumen and aggregates was investigated [24–26]. These studies showed that the SDM is a suitable technique for assessing the effect of moisture on the bitumen-aggregate system.

The bitumen modification with additives is one of the common methods to reduce moisture damage of asphalt compounds [27, 28]. Also, the epoxy resin is a type of polymer, which has many applications in engineering industries. The effect of this additive on improving the performance of bitumen and asphalt compounds has been previously studied [29]. Bitumen modification process is usually accomplished at a high temperature and a specified mixing time. The available studies in the open literature proved that changing the conditions such as mixing time, mixing temperature, and mixing speed during the preparation of the modified bitumen, affected on the performance of the bitumen [30–32]. Pérez-Lepe [33] showed the influence of the conditions during the bitumens modification process with polymer additives on their rheological properties. In another study, the influence of the processing of bitumen modified with tire crumb rubber and polymeric additives was studied [31]. The results showed that a mixing time above 45 minutes exponentially increased the viscosity of the bitumen. Although the influence of the conditions for the preparation of the modified bitumens on their performance has been proven, their effect on the performance of asphalt

compounds in terms of moisture sensitivity, has been rarely studied.

This study focuses on the effect of amount of epoxy resin percentage and its mixing time during the bitumen modification process on the moisture sensitivity of asphalt compounds including two kinds of aggregates. Accordingly, two times, i.e. 10 and 60 minutes as a short and a long mixing times, respectively, are chosen to prepare bitumen modified with epoxy resin. In addition, the SDM is used to evaluate the moisture sensitivity of each asphalt compound by the measuring of moisture sensitivity parameters associated to the surface free energy of bitumen and aggregate.

## 2. SURFACE FREE ENERGY CONCEPT

The surface free energy is defined as the amount of energy required to create a unit surface area of a material under vacuum [34]. The surface free energy of a material ( $\gamma^{total}$ ) consists of two components: a non-polar or Lifshitz-van der Waals component ( $\gamma^{LW}$ ) and a polar component or acid-base component ( $\gamma^{AB}$ ). Consequently, the surface free energy is given by Equation (1) as follows:

$$\gamma^{total} = \gamma^{LW} + \gamma^{AB} \quad (1)$$

The acid-base component ( $\gamma^{AB}$ ) can be calculated according to Equation (2):

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

where ( $\gamma^+$ ) is the Lewis acid parameter of the surface free energy and ( $\gamma^-$ ) is the Lewis base parameter of the surface free energy [35].

**2. 1. Work of Cohesion** The work of cohesion for a material is the required work for splitting a column of the material with a unit cross sectional area into two separate columns. From the definition of surface free energy, it is easy to derive the work of cohesion,  $W_{BB}$ , of the bitumen (material 'B') as follows:

$$W_{BB} = \gamma_B^{total} \quad (3)$$

the higher value of this parameter indicates the more resistant of the bitumen against cohesive failure [15].

## 2. 2. Work of Adhesion and Work of Debonding

The adhesive bond strength under dry conditions refers to the work done by adhesion phase between bitumen and aggregate. The adhesive bond strength in the presence of water is termed the work of debonding. The work of adhesion and the work of debonding can be calculated using Equations (4) and (5), respectively, i.e., from the surface free energy components of the bitumen, aggregate and water. A higher value of the

work of adhesion and work of debonding indicates the more resistance to moisture damage [34].

$$W_{AB} = 2\sqrt{\gamma_A^{LW}\gamma_B^{LW}} + 2\sqrt{\gamma_A^+\gamma_B^-} + 2\sqrt{\gamma_A^-\gamma_B^+} \quad (4)$$

$$W_{AWB}^{wet} = 2\gamma_W^{LW} + 2\sqrt{\gamma_A^{LW}\gamma_B^{LW}} - 2\sqrt{\gamma_A^{LW}\gamma_W^{LW}} - 2\sqrt{\gamma_B^{LW}\gamma_W^{LW}} + 4\sqrt{\gamma_W^+\gamma_W^-} - 2\sqrt{\gamma_W^+(\gamma_A^- + \gamma_B^-)} - 2\sqrt{\gamma_W^-(\gamma_A^+ + \gamma_B^+)} + 2\sqrt{\gamma_A^+\gamma_B^-} + 2\sqrt{\gamma_A^-\gamma_B^+} \quad (5)$$

where  $\gamma^{LW}$ ,  $\gamma^+$  and  $\gamma^-$  are the surface energy components. In addition, A, B, and W denote aggregate, bitumen, and water, respectively.

### 2. 3. Energy Parameter Related to Moisture Sensitivity of Asphalt Compound

The two main factors of asphalt compound stripping are the reduction of bitumen adhesion to aggregates (EP1) and the reduction of bitumen wetting on the aggregate surface (EP2), which they are given by Equations (6) and (7).

$$EP1 = \frac{W_{AB}}{W_{AWB}^{wet}} \quad (6)$$

$$EP2 = \frac{W_{AB} - W_{BB}}{W_{AWB}^{wet}} \quad (7)$$

The greater the energy EP1 parameter, the higher the asphalt compound resistance against moisture [15]. Even though EP1 is an important parameter in determining the stripping of asphalt compound, it does not determine the degree of wettability of aggregates surface by a bitumen, which is represented by EP2 parameter. A higher the EP2 parameter indicates more wettability the bitumen on the aggregate surface, so that the stripping of bitumen-aggregate is less probable [15].

## 3. MATERIALS AND METHOD

In this study, nine types of bitumens were used, namely, eight modified ones at two mixing times and base bitumen, along with two types of aggregate, i.e., siliceous matter and limestone. Different amounts of epoxy resin (2, 4, 6 and 8%) were used to prepare the modified bitumen that is within the range used by Çubuk et al. [36]. As reported in the open literature, epoxy resin, as a bitumen modifier, has also been applied at high percentages, up to 50% [37]. In this study, a lower amount of epoxy resin (2–8%) was utilized to modify bitumen, from the economic point of view [38]. The prolonged mixing time can age bitumen and reduce the performance of bitumen and asphalt compounds containing it, during the modification process. Nevertheless, when bitumen is mixed with additives for a short mixing time, bitumen and additives fail to mix

properly, leading to a weak in bitumen and asphalt pavement structure. Therefore, it is imperative to determine the influence of mixing time and its optimal value for bitumen modification with additives. A minimum mixing time of 10 minutes was chosen to ensure the additive adequately mixes with bitumen, and a maximum 60 minutes was selected to avoid aging bitumen at a constant temperature and speed of mixing condition.

In overall, 18 types of asphalt compounds were evaluated. By measuring the surface free energy of each type of the bitumen and aggregate, the energy parameters associated to the moisture sensitivity of each asphalt compounds were calculated, thereby the moisture resistance of each bitumen and aggregate composition was evaluated. Figure 1 shows the methodology used in this study to obtain the effect of mixing time on adhesion and wetting performances.

### 3. 1. Materials

The commonly used bitumen in Iran which has been provided by Isfahan refinery with 60–70 penetration grade, was used. The main characteristics of the used bitumen are set out in Table 1. Two types of aggregates, i.e. limestone and siliceous, which have a considerable range of characteristics in aspect of mineralogy and degree of moisture resistant variety were also used, here. The chemical compositions of the aggregates that were obtained by performing XRF test are listed in Table 2. The epoxy resin applied in all experiments as the modifying agent was a type of Bisphenol-A Diglycidyl Ether, whose characteristics and chemical open formula are listed in Table 3 and Figure 2, respectively.

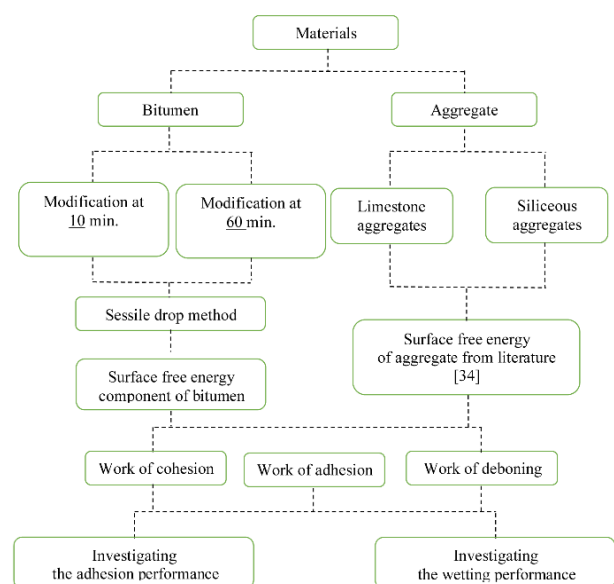


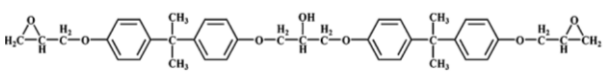
Figure 1. Research plan

**TABLE 1.** Properties of the bitumen used

Characteristic	Standard	Value
Specific gravity at 25 °C, (g/cm <sup>3</sup> )	ASTM D70	1.02
Penetration (100 g, 5 s, 25 °C), 0.1 mm	ASTM D5	65
Softening point (°C)	ASTM D36	50
Flash point (°C)	ASTM D92	324
Kinematic viscosity at 135 °C, (c.st)	ASTM D2170	344
Ductility (25 °C, 5 cm/min), cm	ASTM D113	155

**TABLE 2.** Chemical compositions of aggregates used

Component	Limestone (%)	Siliceous (%)
K <sub>2</sub> O	0.1	0.5
Fe <sub>2</sub> O <sub>3</sub>	0.3	0.4
MgO	1.6	1.0
Al <sub>2</sub> O <sub>3</sub>	1.2	9.0
SiO <sub>2</sub>	1.3	55.0
CaO	52.1	10.0
L.O.I.	43.4	24.1

**Figure 2.** Chemical formula of BPA type epoxy resin**TABLE 3.** Chemical compositions of epoxy resin used

Characteristic	Value
Equivalent weight of epoxy (g/eq)	184-190
Viscosity (cps at 25°C)	11500-13500
Hy-Cl (wt%)	0.05 max
Specific gravity (g/cm <sup>3</sup> at 20°C)	1.17
Color (g)	0.5 max

**3. 2. Bitumen Modification Process** Bitumen modified were prepared by adding epoxy resin percentages being as follows 0, 2, 4, 6, and 8%(w/w) under a specific process. Firstly, pure bitumen preheated at 130 °C was mixed with the specified amount of the mentioned epoxy resin percentages (0, 2, 4, 6, and 8% (w/w) for two mixing times of 10 and 60 minutes at a speed of 600 rpm and temperature of 130 °C. After completing the mixing process, the bitumen samples were kept at 110°C and then 25°C for one hour and one day, respectively. According to this methodology, 8 types of modified bitumens were prepared, at which 4 types refer to mixing time of 10 minutes and 4 remaining ones refer to long mixing time of 60 minutes.

### 3. 3. Sessile Drop Method (SDM) and Measuring Surface Free Energy of Bitumen

SDM is a common method for measuring the surface free energy of solids and liquids [15]. In this study, it was used for measuring surface free energy components of bitumens. According to this method, the surface free energy is calculated using the contact angles of three probe liquids on the bitumen substrate. The probe liquids used in this study were distilled water, ethylene glycol, and formamide. These liquids always have the same magnitude of surface free energy component values, which can also be used to calculate the surface free energy components for other materials, such as bitumen, as reported by Little and Bhasin [15]. The surface free energy components of the mentioned probe liquids for the bitumen are presented in Table 4. In the process of SDM, the prepared bitumens were heated to 135 °C in a 50 ml container. The bitumen heated was poured onto the glass slide surface with dimensions of 75 \* 25 mm. The glass samples covered with bitumen were stored in a desiccator and cooled to room temperature. For each of the bitumens studied, at least 3 samples (the number of probe liquids) were prepared. The bitumen glass slides were placed between a light source and a camera. A micro syringe filled with the probe liquid was placed above the bitumen surface and a small drop of the probe liquid from the syringe was dispensed on a bitumen surface and an image of the drop was captured using a camera (Figure 3). For each bitumen sample, the contact angle at least three drops of each probe liquid were measured and their average was taken as the contact angle of that probe liquid with the bitumen.

The contact angle values are substituted into the following equation to obtain the surface free energy components of bitumen:

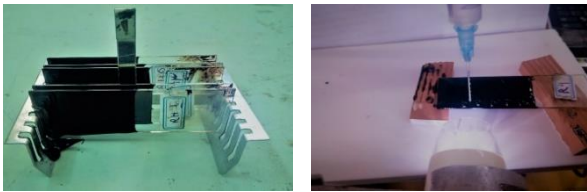
$$\gamma_{Li}(1 + \cos\theta) = 2\sqrt{\gamma_B^{LW}\gamma_{Li}^{LW}} + 2\sqrt{\gamma_B^+\gamma_{Li}^+} + 2\sqrt{\gamma_B^-\gamma_{Li}^-} \quad (8)$$

where  $\gamma_{Li}^{LW}$ ,  $\gamma_{Li}^+$ , and  $\gamma_{Li}^-$  are the surface free energy components of the probe liquid and  $\gamma_B^{LW}$ ,  $\gamma_B^+$ , and  $\gamma_B^-$  are the surface energy components of the bitumen. Furthermore,  $\gamma_{Li}$  is the total surface energy of the probe liquid and  $\theta$  represents the contact angle between the bitumen and the probe liquid. By substituting the surface free energy values of each probe liquid in Equation (8), three equations with three unknowns ( $\gamma_B^{LW}$ ,  $\gamma_B^+$ , and  $\gamma_B^-$ ) are obtained. By solving the set of these equations, the surface free energy components of the bitumen are determined.

**3. 4. Surface Free Energy of Aggregates** Table 5 presents the surface free energy components for both aggregates of limestone and siliceous. It can be seen that siliceous has the higher basic polar component than that of limestone. likewise, acidic polar component and total SFE of limestone are higher than those of siliceous.

**TABLE 4.** Surface free energy components of probe liquids

Probe liquid	Non-polar component ( $\mu\text{J}/\text{m}^2$ )	Acidic polar component ( $\mu\text{J}/\text{m}^2$ )	Basic polar component ( $\mu\text{J}/\text{m}^2$ )	Total surface free energy ( $\mu\text{J}/\text{m}^2$ )
Distilled water	21.8	25.5	25.5	72.8
Ethylene glycol	29.0	1.9	47.0	48.0
Formamide	39.0	2.3	39.6	58.0

**Figure 3.** The SDM procedure**TABLE 5.** Surface free energy values of aggregates used [15]

Aggregate type	Non-polar component ( $\mu\text{J}/\text{m}^2$ )	Acidic polar component ( $\mu\text{J}/\text{m}^2$ )	Basic polar component ( $\mu\text{J}/\text{m}^2$ )	Total surface free energy ( $\mu\text{J}/\text{m}^2$ )
Limestone	44.1	2.4	259.0	93.6
Siliceous	48.8	0	412	48.8

## 4. RESULT AND DISCUSSION

**4.1. Contact Angle Measurement** The measured contact angles for different probe liquids, i.e., distilled water, ethylene glycol and formamide, on the bitumen surface are shown in Figure 4(a, b and c). Figure 4 presents the standard deviations by error bars for the bitumen specimens with three probe liquids. For each probe liquid and asphalt binder tested in five replicates, there is a small deviation less than  $5^\circ$  in contact angle [15].

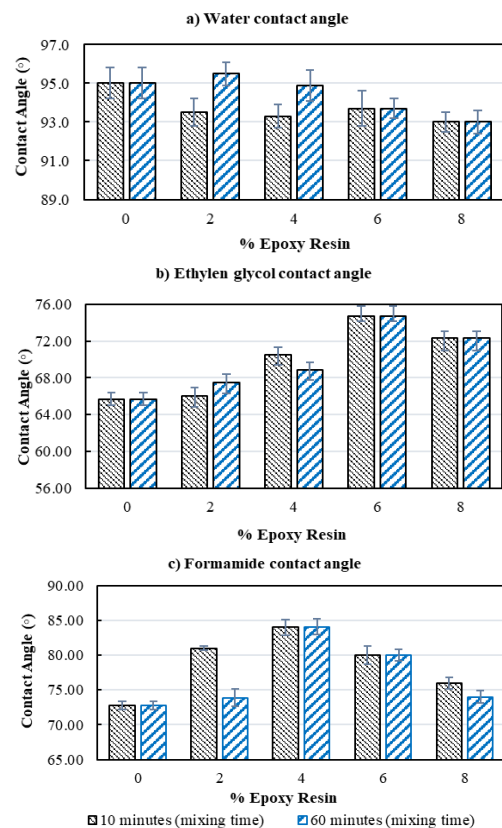
In addition, the findings show the maximum standard deviation of  $1.3^\circ$  that confirms the repeatability of results. According to the figure, for the probe liquids of distilled water and ethylene glycol, the contact angles of bitumens modified with 2 and 4% epoxy resin are different in mixing time of 10 and 60 minutes; however, the contact angles of bitumens containing 0, 6, and 8% epoxy resin are equal for both mixing times of 10 and 60 minutes. The contact angle of the formamide probe liquid for mixing time of 10 minutes is higher than that of 60 minutes for bitumens modified by 2 and 8% epoxy resin; however, these values are equal for the bitumens containing 4 and 6% epoxy resin for both mixing times. According to the results, the change of the process

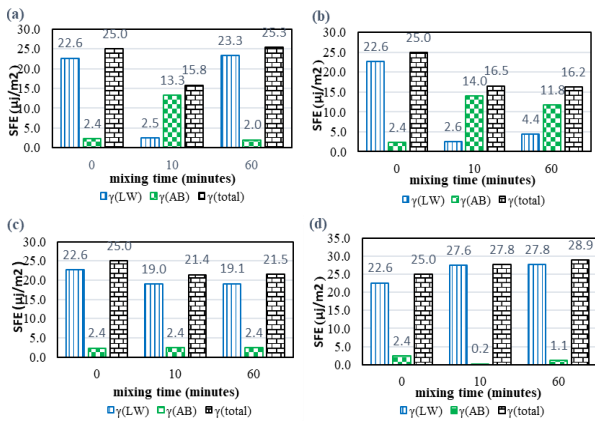
mixing time and the percentage of epoxy resin in bitumen leads to a change of the contact angle of the probe liquids with the bitumen surface due to a change in the chemical nature of bitumen.

## 4.2. Surface Free Energy Component of Bitumens

The surface free energy components of each bitumen were obtained using the contact angle measured with the probe liquids by SDM. Figure 5 shows the surface free energy components of bitumens modified with 2, 4, 6 and 8% epoxy resin and mixed for different mixing times (10 and 60 minutes). The zero-minute mixing time shown in Figure 5 refers to the value of the surface free energy components for the base bitumen. As can be seen, the surface free energy components change with the epoxy resin percentage and mixing time. The values of the surface free energy components are 25, 2.4, and  $22.6 \mu\text{J}/\text{m}^2$ , respectively, for the total, polar, and non-polar surface free energy components in the case of base bitumen. These results mainly show the base bitumen, unmodified, is a kind of the non-polarity material.

The bitumen with 4% epoxy resin shows the highest polarities of 14 and  $11.8 \mu\text{J}/\text{m}^2$  for the mixing times of 10 and 60 minutes, respectively, among the other cases. Whereas the bitumen modified with 8% epoxy resin exhibits the maximum value of total surface free energy

**Figure 4.** Measurement of contact angle of probe liquids with bitumen



**Figure 5.** Surface free energy components for modified bitumens at different mixing times: a) 2% epoxy resin, b) 4% epoxy resin, c) 6% epoxy resin, d) 8% epoxy resin

and the non-polar surface free energy, it shows the minimum value of polar surface free energy. On the other hand, the influence of mixing time is quite different for bitumens with various epoxy resin percentages. In the surface free energy component of the bitumen containing 2% epoxy resin and mixed for 10 minutes, the total surface free energy and the non-polar surface free energy decrease; while, the polarity of the modified bitumen increases in comparison with the base bitumen. When the mixing time is 60 minutes, the surface free energy components of the modified bitumen are very close to those of base bitumen surface free energy component. These results show that at 2% epoxy resin, 10 minutes mixing time has a greater effect on the polarity of the modified bitumen than 60 minutes mixing time. There is an increase in the polar component of the surface free energy for the bitumen modified with 4% epoxy resin comparison with the base bitumen. It should be noted that modified bitumens for 10 minutes mixing time lead to more polarity than those mixed for 60 minutes. In the case of bitumens containing 6% epoxy resin, the total surface free energy and the non-polar surface free energy decrease compared to the base bitumen, while, the polar surface free energy does not change. In fact, the value of polarity of this type of modified bitumen is similar to the base bitumen independently of the mixing time. Regarding the bitumen containing 8% epoxy resin, the amount of total surface free energy and the non-polar surface free energy increased compared to the base bitumen, but the polar surface free energy decreased. Therefore, the results show that the effect of mixing time on the bitumen surface free energy components for the bitumens containing lower percentages of epoxy resin (2 and 4%) is more noticeable than the bitumens containing higher percentages, i.e. 6 and 8%.

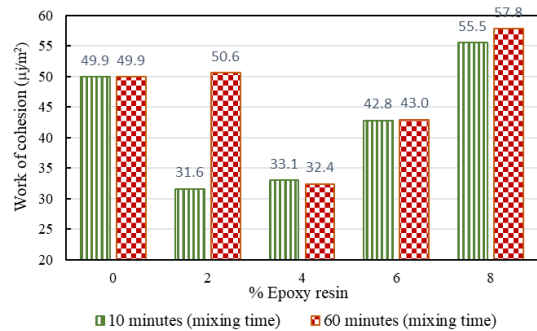
**4. 3. Work of Cohesion of Bitumens**

As mentioned before, the cohesion energy or work of

cohesion is calculated from the surface free energy components of the bitumen using Equation (3). It should be noted that when the value of this parameter is high, the bitumen is more resistant against crack propagation and crack growth [15]. Figure 6 shows the cohesion energy for different modified bitumens. As observed, adding epoxy resin to bitumen at both mixing times alters the work of cohesion of bitumen. The bitumens modified with 4 and 6% epoxy resin for both mixing times, and with 2% epoxy resin for the mixing time of 10 minutes, decrease the work of cohesion compared to the base bitumen. When bitumen is mixed with 8% epoxy resin for 10 and 60 minutes, and also with 2% epoxy resin for 60 minutes, its cohesive performance has improved compared to the unmodified bitumen. Furthermore, the maximum and minimum values of the work of cohesion are determined by mixing of bitumen with 8% epoxy resin for 60 minutes and with 2% epoxy resin for 10 minutes, respectively. It is also found that the values of work of cohesion for the 4 and 6% epoxy resin modified bitumens are almost the same for both mixing times. Moreover, the work of cohesion improved by 4.1% for the 8% epoxy resin modified bitumen prepared at a longer mixing time. It is also observed that the work of cohesion increased by 60% for the 2% epoxy resin modified bitumen prepared at a longer mixing time. In overall, the results show that the work of cohesion is affected by the mixing time of bitumen with epoxy resin based on the amount of additive used.

**4. 4. Work of Adhesion and Debonding**

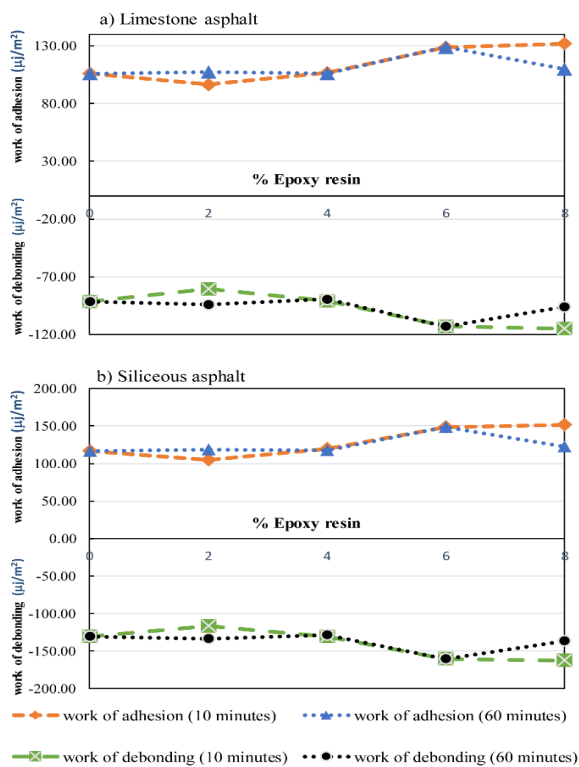
Figure 7 shows the variation of the values of work of adhesion and work of debonding for two mixing times of 10 and 60 minutes. Increasing the values of both parameters in positive direction shows the enhancing the performance of the bitumen-aggregate system against moisture. According to Figure 7a (limestone asphalt compounds) and Figure 7b (siliceous asphalt compounds), although the effect of mixing time on these parameters is negligible for bitumens containing 4 and 6% epoxy resin, the corresponding effect for bitumens containing 2 and 8% epoxy resin is considerable. These results show that



**Figure 6.** Work of cohesion for bitumens modified for 10 and 60 minutes mixing times

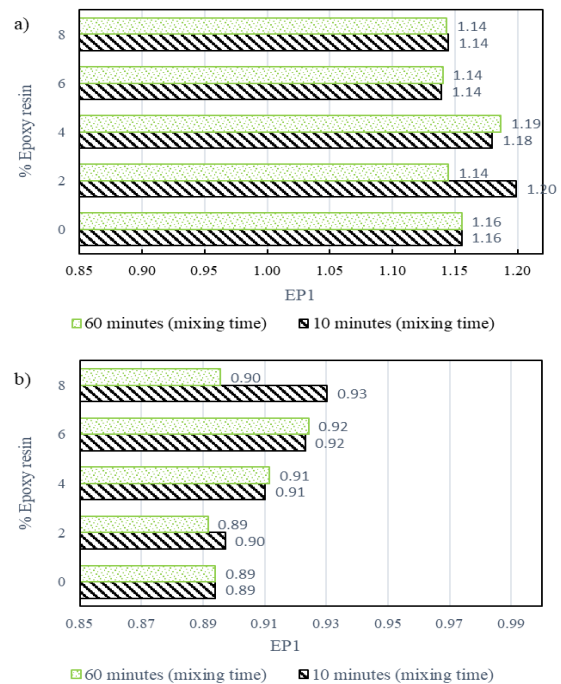
the effect of mixing time on the mentioned parameters depends on the percentage of additive, epoxy resin, in bitumen. As observed, both asphalt compounds (limestone and siliceous) have a similar impact on the work of adhesion and work of debonding based on the amount of epoxy resin in modified bitumen. The work of adhesion and work of debonding and thereby the performance of asphalt compound decrease with longer mixing time for the bitumen modified with 2% epoxy resin, whereas the performance of asphalt compound improves with time for the bitumen modified with 8% epoxy resin. According to Equations (6) and (7), as discussed in section 4.5, these results were used to calculate the magnitude of energy parameters related to moisture resistance of asphalt compound.

**4. 5. Energy Parameters Related To Moisture Resistance of Asphalt Compound** EP1 and EP2 are the parameters that predict the moisture resistance of the asphalt compounds. EP1 and EP2 denote the bitumen adhesion to the aggregate and the extent of bitumen wetting on the aggregate surface, respectively. It should be noted, when EP1 and EP2 increase, the moisture resistance of bitumen-aggregate also increases. In the following, the behavior of these two parameters for the asphalt compounds containing different aggregates are studied for two mixing times.



**Figure 7.** Work of adhesion and work of debonding for limestone and siliceous asphalt compounds for 10 and 60 minute mixing times

**4. 5. 1. Influence of Mixing Time on EP1** Figure 8 shows the variation of EP1 for asphalt compounds containing limestone/siliceous aggregates and modified bitumen for mixing times of 10 and 60 minutes. The minimum and maximum values of EP1 parameter for limestone asphalt are 1.14 and 1.20, respectively, while, the corresponding values for asphalt compounds containing siliceous aggregate are 0.89 and 0.93. These results of EP1 parameter show that asphalt compounds comprising limestone aggregate leads to a better adhesion performance than that of the siliceous aggregate. It is also evident that the effect of mixing time on the EP1 parameter is insignificant. Nevertheless, for the mixing time of 10 minutes, the value of EP1 increases by a maximum 5.26% in comparison with the mixing time of 60 minutes for the asphalt compound containing limestone aggregate and bitumen modified with 2% epoxy resin. As observed, in the case of siliceous compounds, the maximum difference between the values of EP1 occurs for the modified bitumen with 8% epoxy resin under both mixing times of 10 and 60 minutes. Also, the EP1 parameter for the prepared bitumen with a mixing time of 10 minutes is 3.3% higher than that of 60 minutes. Consequently, the mixing time of the bitumen with different percentages of epoxy resin cannot be an effective parameter for improving the adhesive performance of the asphalt compounds including limestone/siliceous aggregates and modified bitumen. Also, an increase of the mixing time not only is



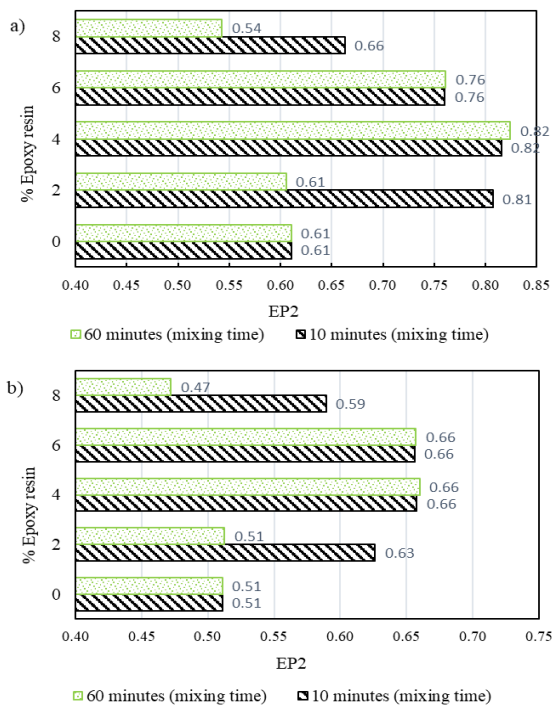
**Figure 8.** EP1 variations for asphalt mixtures containing modified bitumen using mixing times of 10 and 60 minutes: a) limestone aggregate, b) siliceous aggregate

ineffectual in improving the asphalt compound performance, but also leads to energy loss in the process. Accordingly, a mixing time of 10 minutes is sufficient to reach a suitable adhesive performance.

**4. 5. 2. Influence of Mixing Time on EP2**

Figure 9 shows the variation of EP2 when siliceous or limestone aggregates are mixed with each modified bitumen for mixing times of 10 and 60 minutes. As shown in this figure, the value of EP2 for limestone aggregate is greater than that of the siliceous aggregate for the asphalt compounds with almost all percentages of epoxy resin. This reveals that the wetting performance of limestone aggregates is higher than that of siliceous aggregates. For both aggregates in the compounds, the mixing time of 10 minutes for preparing the modified bitumens with 2 and 8% epoxy resin leads to higher values of EP2 than those preparing the modified bitumens for 60 minutes' mixing time. This finding proves the importance of the mixing time impact on the process of providing the modified bitumens, thereby on the performance of asphalt compounds. Therefore, an appropriate mixing time of modified bitumen beside the proper value of additive percentage adjust the wetting performance of the asphalt compound.

When 4 and 6% epoxy resin are used to modify the bitumen, the samples prepared using the both mixing times leads to the identical EP2 values for both



**Figure 9.** EP2 variations for asphalt mixtures containing modified bitumen using mixing times of 10 and 60 minutes: a) limestone aggregate, b) siliceous aggregate times

aggregates. However, there are significant differences between the values of EP2 for both mixing times and aggregates in the cases of the bitumens modified by 2 and 8% epoxy resin. The values of EP2 increase about 32.8 and 23.5% for the mixing time of 10 minutes in comparison with those of mixing time of 60 minutes for the modified bitumens with 2% epoxy resin mixed with limestone and siliceous aggregates, respectively. Moreover, 8% epoxy resin with limestone and siliceous aggregates at the mixing time of 10 minutes led to increasing in the values of EP2 of 22.2 and 25.5%, respectively, compared with those of 60 minutes mixing time. Generally, an increase in the mixing time of modified bitumen causes a decrease in the wetting performance of asphalt' compounds, so that the mixing time of 10 minutes is a better time than 60 minutes.

**5. CONCLUSIONS**

The effects of mixing time and additive percentage in the modified bitumen with combination of two important aggregates on the moisture sensitivity of asphalt compounds were investigated using surface free energy measurement. The main findings of the study can be summarized as follows:

- The values of total, polar, and non- polar surface free energy for the base bitumen are 25, 2.4 and 22.6  $\mu\text{J}/\text{m}^2$ , respectively; which reveals the bitumen is a kind of non-polarity material.
- The 8% epoxy resin modified bitumen produces the maximum values of total surface free energy and non-polar surface free energy. However, the minimum value of polar surface free energy is also relevant to this specimen. Also, the maximum polarity is obtained by the bitumen modified with 4% epoxy resin with the values of 14 and 11.8  $\mu\text{J}/\text{m}^2$  for the mixing times of 10 and 60 minutes, respectively.
- The effect of mixing time on bitumen surface free energy components is more noticeable in case of bitumens with the lower percentages of epoxy resin additive (2 and 4%) compared to the bitumens with higher ones (6 and 8%).
- In each modified bitumen, the value of the work of cohesion corresponding to the mixing time of 60-minutes is higher than that corresponding to 10-minutes one. In fact, an increase in the mixing time leads to enhancing the cohesion bitumen energy.
- The mixing time affects the work of adhesion and work of debonding based on the percentage of additive in bitumen. There is no important difference between the values of these parameters in the two mentioned mixing times for bitumens containing 4 and 6% epoxy resin. Nevertheless, the values of the work of adhesion and work of debonding for bitumens with the addition of 2 and 8% epoxy resin



for 10- and 60-minutes mixing time are quite different.

- The minimum and maximum values of EP1 parameter for limestone asphalt are 1.14 and 1.20, respectively. Those values for asphalt compounds containing siliceous aggregate are 0.89 and 0.93. These results show that asphalt compounds containing limestone aggregates lead to a better adhesion performance than those containing siliceous aggregates.
- The value of EP2 for limestone aggregates is greater than that of siliceous aggregates for asphalt compounds containing epoxy resin in almost all percentages. This means that limestone aggregates have a higher wetting performance than siliceous aggregates.
- In the modified bitumens with amount of epoxy resin 4 and 6%, the values of EP1 and EP2 parameters are very close to each other for both 10- and 60-minutes mixing times. This means that the mixing time of bitumen with various percentages of epoxy resin is not influential in improving the adhesion and wetting performance of asphalt compounds.
- The asphalt compounds containing bitumens with 2 and 8% epoxy resin prepared for 10 minutes' mixing time show a better performance in terms of adhesion (EP1) and wetting (EP2) compared to those prepared for mixing time 60 minutes.
- Finally, the mixing time of 10 minutes is recommended as a suitable value for bitumens with 0 to 8% epoxy resin in order to reduce the moisture sensitivity of asphalt compounds.

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### Persian Abstract

#### چکیده

اصلاح قیر، امکان بهبود خواص آن را فراهم می‌کند و می‌تواند به عنوان یک روش مناسب در جهت بهبود خواص ترکیب‌های آسفالتی مورد استفاده قرار گیرد. بنابراین این مطالعه به بررسی اثر زمان اختلاط قیرهای اصلاح شده با درصد‌های متفاوت رزین اپوکسی از صفر تا ۸ درصد وزنی قیر، بر حساسیت رطوبتی ترکیب‌های آسفالتی حاوی دو نوع سنگدانه آهکی و سیلیسی پرداخته است. به این منظور، روش قطره چسبیده مورد استفاده قرار گرفت و پارامترهای حساسیت رطوبتی مربوط به انرژی آزاد سطحی شامل اجزای انرژی آزاد سطحی، انرژی پیوستگی، انرژی چسبندگی، انرژی شکست پیوند، میزان ترشوندگی قیر بر سطح سنگدانه، و میزان چسبندگی قیر به سنگدانه محاسبه و مورد ارزیابی قرار گرفتند. نتایج نشان دادند میزان اثر زمان اختلاط قیرهای اصلاح شده با رزین اپوکسی بر حساسیت رطوبتی ترکیب‌های آسفالتی به طور قابل توجهی وابسته به درصد رزین اپوکسی موجود در قیر است. برای قیرهای با ۴ تا ۶ درصد اپوکسی رزین، عملکرد چسبندگی و ترشوندگی قیر به سنگدانه برای زمان‌های اختلاط ۱۰ دقیقه و ۶۰ دقیقه مشابه بود. با این حال، در ترکیب‌های آسفالتی حاوی ۲ و ۸ درصد رزین اپوکسی، قیرهای اصلاح شده با زمان اختلاط ۱۰ دقیقه عملکرد بهتری نسبت به قیرهای با زمان اختلاط ۶۰ دقیقه از خود نشان دادند. اثر زمان اختلاط قیرهای اصلاح شده با رزین اپوکسی بر عملکرد قیر و ترکیب آسفالتی وابسته به درصد افزودنی به کارگرفته شده در قیر است.