Research Article

The Effect of Aggregate-Forming Minerals on Thermodynamic Parameters Using Surface Free Energy Concept and Its Relationship with the Moisture Susceptibility of Asphalt Mixtures

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Moisture damage is known to be a key factor influencing the durability of asphalt pavements. This phenomenon will reduce the mechanical properties of asphalt mixtures due to its destructive effects on the adhesion of the bitumen-aggregate system and the cohesion of the bitumen membrane. The mineral properties of the aggregates of asphalt mixtures are one of the factors affecting moisture susceptibility. Therefore, the surface free energy (SFE) method, as one of the thermodynamic methods, and indirect tensile strength test (ITST) have been used to determine the failure mechanism and the rate of change of adhesion properties of asphalt mixtures under the influence of mineralogical characteristics. Prior to the above tests, an X-ray fluorescence spectrometry (XRF) test was performed to identify aggregate-forming minerals in eight different types of aggregates with various minerals from eight mines, and also the apparent bitumen film thickness was measured. XRF test results showed that most parts of the aggregates in mines 1–6 were composed of SiO₂, which had strong acidic properties, according to the SFE results. ITST results showed that samples constructed with aggregates of mine 7 and 8 (with high CaO mineral content) experienced a higher TSR than other mixtures in all freeze-thaw (F-T) cycles. Results of the SFE method showed that aggregates with more calcareous properties had a smaller acidic component and a larger basic component. In mines 7 and 8, where the nonpolar component of their aggregates was larger, the tendency of the aggregates to adhere and wettability by water decreased. The adhesion free energy (AFE) between bitumen and aggregates with higher CaO and less SiO₂ amounts was more than AFE between bitumen and aggregates with lower CaO and higher SiO₂ amounts, in dry conditions. The presence of higher amounts of CaO in aggregates increased AFE of bitumen-aggregate in presence of water from a negative value to zero. In linear regression analysis, due to the positive regression coefficient of the CaO mineral, this mineral had a positive impact on TSR; on the other hand, due to the negative regression coefficient of the SiO₂ mineral, this mineral had a negative impact on the TSR of asphalt mixtures.

1. Introduction

Moisture damage is one of the factors that reduce the lifespan of asphalt pavements due to the presence of water in it and, as a result, intensifies other damages such as fatigue, rutting, pothole, and so on in pavements, especially in rainy areas [1–3]. In general, the presence of moisture in pavement causes the bitumen membrane to separate from the surface of the aggregate, resulting in a type of failure that engineers call stripping [4, 5]. The main and the most serious consequence of stripping is the loss of strength and integrity of the pavement, which by moving the bitumen over the aggregate surface or breaking the bitumen membrane creates a greater tendency in aggregates to absorb water than their tendency to be covered by bitumen [6–8]. The properties of aggregates are very important in moisture damage, but
negligible study of aggregates in moisture damage has been done. The relationship between the minerals that make up the aggregates and the surface free energy (SFE) can also help in choosing more resistant aggregates. In SFE methods, it is difficult to perform experiments. But if the relationship between the aggregate-forming minerals and the SFE components is roughly determined, it can be concluded that each of the aggregates with its mineral structure has components of SFE. 

Most existing experiments use the comparison of the mechanical performance of mixtures in dry and wet conditions to determine their susceptibility and the potential for moisture-induced damage in asphalt mixtures [9, 10]. Despite the breadth of these experiments, such as the modified Lattman method (AASHTO T283), these methods do not examine the basic characteristics of substances that are effective in the event of moisture-induced damage and cannot explain the reason of the weakness or strength of asphalt mixture performance and offer a suitable corrective solution to enhance the performance of the asphalt mixture when exposed to moisture [11]. Based on this, researches have been conducted in the last two decades to determine moisture susceptibility in asphalt mixtures according to the basic characteristics of substances that are effective in bitumen cohesion and bitumen-aggregate adhesion.

1.1. Literature Review. Moisture is one of the main causes of the deterioration of flexible pavements, and moisture-induced failure can be defined as a decrease in the mechanical properties of the asphalt mixture due to water presence. Moisture damage was first identified in the early 1960s as a major issue; however, in 1980, the damage came to the attention of pavement organizations and industries. Today, despite advances and developments in the mix design of asphalt mixtures and a better understanding of the mechanisms of moisture-induced damage, this damage is still one of the most common and complex problems facing the pavement community [11, 12].

Since 1932, the interface relationships between aggregate and bitumen have been studied in the presence of water [13]. Elphingstone was the first to show that measuring SFE could be utilized as a good tool for predicting fatigue cracking and moisture-induced damage in asphalt mixtures [14]. Cheng in his study on the concepts of SFE, its measurement, and application in asphalt mixtures has indicated that thermodynamic variations in adhesion and cohesion SFE were directly related to the separation at the bitumen-aggregate contact surface and the occurrence of cracks in the bitumen, respectively [15]. Bhasin in his research first developed methods to calculate bitumen and aggregate SFE components. Next, the relationship between thermodynamic parameters, which were obtained from the measurement of aggregate and bitumen SFE components, and the potential of moisture susceptibility in asphalt mixtures were investigated [16]. Howson also explored the use of SFE to identify the potential of asphalt mixtures against moisture [17]. In a study by Hamedi and Nejad, using the results of moisture sensitivity tests of various compositions of asphalt mixtures, the relationship between the potential of moisture-induced damage and thermodynamic parameters was investigated [18]. Arabani and Hamedi showed that thermodynamic parameters, including bitumen cohesion free energy, bitumen coverage on the aggregate surface, adhesion free energy (AFE) of bitumen aggregate, and detachment energy (DE) of the system in the stripping event, could appropriately estimate the moisture sensitivity of various asphalt mixtures [19]. However, Mercado considers the ingredients of asphalt mixtures, including bitumen and aggregates, to be internal factors affecting moisture-induced damage [20].

In various studies, the modification of bitumen properties by the use of anti-stripping additives to increase the strength of asphalt mixtures has been investigated [21–27]. Many other studies have also examined the properties of aggregates. Improving the properties of aggregates using additives is one of the main common techniques to enhance the performance of asphalt mixtures against moisture susceptibility [28–30]. The mineral and chemical composition of an aggregate is considered as an important factor in the susceptibility of asphalt pavement to stripping. They affect surface energy as well as the chemical reaction and are also effective for the absorption coating on the aggregate surface [31, 32]. Asphalt absorption by pavement aggregates is one of the factors that have important consequences for asphalt-aggregate bonding. Many studies have attempted to find a correlation between the physical and chemical properties of aggregates and the asphalt absorption rate. Higher water absorption aggregates are more likely to absorb asphalt than those that have less water absorption [33].

1.2. Problem and Purpose. Aggregate is one of the main decisive factors of the occurrence of moisture-induced damage. Several properties of aggregate affect the moisture susceptibility of asphalt mixtures, such as shape and size, surface texture, fracture, porosity, specific surface area, cleanliness, moisture content, mineral composition, and surface energy [34]. Of course, not all of these features are independent of each other. For example, the composition of minerals affects the SFE components of aggregates.

The composition of the minerals that make up aggregates based on their hydrophilic or hydrophobic properties and their relationship with bitumen is also important in the event of moisture damage. High-carbonate aggregates (hydrophobic or basic aggregates), such as limestone, are easier to coat. This type of aggregate creates a more powerful bonding with the bitumen than the aggregate with a great percentage of silica (acidic or hydrophilic aggregates), which is the main reason for the acidity of the bitumen [35, 36]. The main significant goals of the study are

(i) Investigating the impact of the type of aggregate-forming minerals on aggregate SFE components
(ii) Investigating the effect of the type of aggregate-forming minerals on aggregate-bitumen adhesion
(iii) Investigating the impact of the type of aggregate-forming minerals on the tendency to the stripping phenomenon from a thermodynamic point of view
(iv) Investigating the impact of the type of aggregate-forming minerals on the moisture susceptibility of asphalt mixtures

2. SFE Theory

Several theories describe SFE of substances according to their molecular structure. Two of the main theories are the acid-base theory and the two-component theory. In a study, Fowkes suggested that the total SFE of substances consists of two main components, and divided them into scattered forces, such as nonpolar forces within a molecule including Lifshitz-van der Waals, and forces related to special interactions, including hydrogen bonding, and stated that the total SFE or surface tension is a linear combination of these interactions [37, 38]:

\[ \Gamma_{\text{total}} = \Gamma_{\text{LW}} + \Gamma_{\text{AB}}. \]  

in which \( \Gamma_{\text{total}} \) presents the total SFE of substances, \( \Gamma_{\text{AB}} \) presents the SFE polar component, and \( \Gamma_{\text{LW}} \) presents the SFE nonpolar component. This model is known as the two-component theory, and other theories that have been extensively applied to describe the SFE components of various substances include acid-base theory [39], in which the total SFE of any substance is classified into components of the nonpolar, the Lewis acid, and the Lewis base, based on the type of forces of surface of molecular. The total SFE is as

\[ \Delta G_{\text{i}12} = \Gamma_{12} - \Gamma_{13} - \Gamma_{23} = -2 \left[ \Gamma_3^{\text{LW}} + 2 \sqrt{\frac{\Gamma_1^{\text{LW}}}{\Gamma_3^{\text{LW}}}} - \sqrt{\frac{\Gamma_1^{\text{LW}}}{\Gamma_3^{\text{LW}}}} \frac{\Gamma_1^{\text{LW}}}{\Gamma_3^{\text{LW}}} - \sqrt{\frac{\Gamma_1^{\text{LW}}}{\Gamma_3^{\text{LW}}}} - \sqrt{\frac{\Gamma_1^{\text{LW}}}{\Gamma_3^{\text{LW}}}} - \sqrt{\frac{\Gamma_1^{\text{LW}}}{\Gamma_3^{\text{LW}}}} - \sqrt{\frac{\Gamma_1^{\text{LW}}}{\Gamma_3^{\text{LW}}}} - \sqrt{\frac{\Gamma_1^{\text{LW}}}{\Gamma_3^{\text{LW}}}} \right]. \]  

Using equations (1)–(5), the SFE of the components of the asphalt mixture, i.e., bitumen and aggregate, as well as the amounts of SFE for the whole mixture are calculated.

3. Materials

3.1. Aggregates. Eight aggregate types with different characteristics from Iranian mines, according to Figure 1, were used to investigate the effect of the minerals that make up aggregates on moisture susceptibility. An X-ray fluorescence spectrometry (XRF) test was used to determine the constituent minerals. The grading of the aggregates in the research was according to the ASTM standard, which specifies that the nominal and maximum size of the aggregates should be 12.5 mm and 19 mm, respectively, and it is illustrated in Figure 2. Table 1 also indicates the physical features of the aggregates.

3.2. Bitumen. In this research, bitumen with a penetration grade of 60–70 prepared from the Jey oil refinery was used, the features of which are indicated in Table 2.

\[ \Gamma_{\text{total}} = \Gamma_{\text{LW}} + \Gamma_{\text{AB}} = \Gamma_{\text{LW}} + 2\sqrt{\Gamma^- \Gamma^+}, \]  

where \( \Gamma^- \) represents the acidic component and \( \Gamma^+ \) represents the basic component. The polar component consisting of the Lewis base and the Lewis acid components as given in

\[ \Gamma_{\text{AB}} = 2 \sqrt{\Gamma^- \Gamma^+}. \]  

The AFE (\( \Delta G_{\text{AB}}^i \)) consists of polar and nonpolar components. The AFE between the aggregate and bitumen is determined as

\[ \Delta G_{\text{AB}}^i = \Delta G_{\text{AB}}^{\text{LW}} + \Delta G_{\text{AB}}^{\text{LW}} = -2 \left[ \sqrt{\frac{\Gamma_1^{\text{LW}}}{\Gamma_3^{\text{LW}}}} + \sqrt{\frac{\Gamma_1^{\text{LW}}}{\Gamma_3^{\text{LW}}}} + \sqrt{\frac{\Gamma_1^{\text{LW}}}{\Gamma_3^{\text{LW}}}} \right]. \]  

where \( \Delta G_{\text{AB}}^{\text{LW}} \) and \( \Delta G_{\text{AB}}^{\text{LW}} \) are nonpolar and polar components of AFE; \( \Gamma_1^{\text{LW}}, \Gamma_1^{\text{LW}}, \Gamma_1^{\text{LW}}, \) and \( \Gamma_1^{\text{LW}}, \Gamma_1^{\text{LW}}, \Gamma_1^{\text{LW}}, \) and \( \Gamma_1^{\text{LW}}, \Gamma_1^{\text{LW}}, \Gamma_1^{\text{LW}}, \) present the aggregate SFE components.

Equation (5) is applied to measure the AFE of aggregate-bitumen in the presence of water, with the indexes of 1, 2, and 3, which indicate asphalt, aggregate, and water, respectively. If the AFE amounts are negative, meaning that the two substances incline to detach from one another, the more negative the amount, the higher the inclination.

4. Testing Plan

In this research, 8 different aggregate types were used. The reason for using different aggregates was to be able to study a wide range of minerals. A common type of bitumen (bitumen with a penetration grade of 60–70) was used. In order to eliminate the effect of the bitumen type, one type of bitumen was used in all cases. In fact, the optimal bitumen percentage was determined. Then, for each asphalt mixture, minerals that make up all 8 types of aggregates were identified by the XRF test. In order to simulate environmental conditions, the samples were subjected to freeze-thaw (F-T) cycle numbers of 1, 3, and 5, according to AASHTO T283 standard, using the indirect tensile strength test (ITST). Finally, the SFE method was applied to investigate the adhesion properties of aggregate and bitumen constituted from different minerals. The testing plan of the present study is in accordance with Figure 3.
4.1. X-Ray Fluorescence Spectroscopy (XRF) Test. One of the important parameters in determining the moisture susceptibility of different aggregates is the structure of aggregate minerals. The XRF test was used to determine the structure of the aggregates applied. The XRF is a device analysis method that uses the X-ray diffraction spectrum method to decompose surface layers. This device has the ability to perform elemental analysis quantitatively and qualitatively of mineral samples such as geological samples, minerals, rocks, glass, cement, ceramics, metal alloys, and so on. Stimulation of the sample due to X-ray radiation causes electron transfer in different layers of the atom, in which
Table 1: The physical features of the aggregates.

| Features                          | Standard | Mines |
|----------------------------------|----------|-------|
|                                   | 1  | 2  | 3  | 4  | 5  | 6  | 7  | 8  |
| Specific gravity of coarse aggregate |      |     |    |    |     |    |    |    |
| Effective                         | 2.56 | 2.46 | 2.58 | 2.59 | 2.53 | 2.48 | 2.61 | 2.59 |
| Bulk                              | 2.57 | 2.49 | 2.59 | 2.61 | 2.55 | 2.49 | 2.63 | 2.60 |
| Apparent                          | 2.59 | 2.50 | 2.62 | 2.63 | 2.58 | 2.52 | 2.65 | 2.62 |
| Specific gravity of fine aggregate |      |     |    |    |     |    |    |    |
| Effective                         | 2.54 | 2.44 | 2.55 | 2.55 | 2.50 | 2.46 | 2.60 | 2.57 |
| Bulk                              | 2.57 | 2.47 | 2.57 | 2.57 | 2.52 | 2.48 | 2.62 | 2.58 |
| Apparent                          | 2.59 | 2.48 | 2.60 | 2.59 | 2.53 | 2.49 | 2.65 | 2.61 |
| Specific gravity (filler)         |      |     |    |    |     |    |    |    |
|                                   | 2.56 | 2.43 | 2.55 | 2.53 | 2.49 | 2.44 | 2.55 | 2.56 |

Table 2: The features of bitumen in the study.

| Features                          | Standard | Bitumen |
|----------------------------------|----------|---------|
| Penetration grade (0.1 mm)       | ASTM: D5 | 66      |
| Ductility (cm)                   | ASTM: D113 | 112      |
| Specific gravity at 25°C         | ASTM: 70–76 | 1.02    |
| Softening point (°C)             | ASTM: D36 | 51      |
| Trichloroethylene solubility (%) | ASTM: D2042 | 99.5    |
| Loss on heating (%)              | ASTM: D1754 | 0.75  |
| Flash point (°C)                 | ASTM: D92  | 262     |

Figure 3: Testing plan of the present study.
each electron transfer is accompanied by the emission of a spectral X-ray line. The wavelength of the emitted spectral lines is the basis for the qualitative analysis of the elements and the intensity of the rays in proportion to the frequency or quantity of the elements in the sample.

4.2. Marshall Mix Design. The Marshall mix design in this study was applied according to AASHTO T245 to specify the optimal bitumen percentage.

4.3. Bitumen Apparent Film Thickness on Aggregate Surface. The bitumen membrane in the asphalt mixture means the average thickness of the bitumen on the surface of the aggregates, which is called bitumen apparent film thickness (AFT). Some studies have illustrated that AFT is another important factor in the occurrence of moisture-induced damage, as this factor affects the durability of the asphalt mixture [40, 41]. The asphalt mixture in which AFT is low is brittle and is not durable enough, while the asphalt mixture with a high bitumen thickness is prone to rutting and shrinkage. The high thickness of the bitumen membrane produces a durable asphalt mixture. In fact, it decreases the water moving possibility in the asphalt mixture [42], which decreases the severity of moisture-induced damage in asphalt mixtures [43]. Whether the rupture caused by moisture is adhesion or cohesion also depends on the thickness and the type of bitumen [44]. In this study, to obtain AFT, the method provided in the NCHRP 567 report, according to equation (6) has been used, in which, the numerator presents the volume of bitumen consumed per unit volume of asphalt mixture, and its denominator indicates the surface of aggregates per unit volume of asphalt mixture [45].

\[
AFT = \frac{1000 \times VBE}{S_s \times P_s \times G_{mb}},
\]

where AFT is the bitumen apparent film thickness (μm), VBE is the percentage of effective bitumen volume relative to the total volume of asphalt mixture (%), \( P_s \) presents the percentage of the weight of aggregates to the total weight of the mixture (%), \( S_s \) presents the apparent specific surface area of aggregates (m²/kg), and \( G_{mb} \) presents specific volumetric weight of the mixture (kg/m³). Equation (7) was used to achieve \( S_s \):

\[
S_s = \left(\frac{1}{1000G_{mb}}\right) \left[1.4(P_{50} - P_{37.5}) + 2.0(P_{37.5} - P_{25}) + 2.8(P_{25} - P_{19.5}) + 3.9(P_{19.5} - P_{12.5}) + 5.5(P_{12.5} - P_{9.5}) + 8.9(P_{9.5} - P_{7.5}) + 17.9\right.
\[
\left.\times 36(P_{4.75} - P_{2.36}) + 36(P_{2.36} - P_{1.18}) + 71.3(P_{1.18} - P_{0.6}) + 141(P_{0.6} - P_{0.3}) + 283(P_{0.3} - P_{0.15}) + 566(P_{0.15} - P_{0.075}) + 1600(P_{0.075})\right]\]

where \( G_{mb} \) represents the total volumetric weight of aggregates (kg/m³) and \( P_s \) is the passing percentage through a sieve with a diameter of x mm. This apparent specific surface is part of the surface of the aggregates on which the thickness of the bitumen membrane is effectively formed [45].

4.4. Indirect Tensile Strength Test. In this study, AASHTO T283 standard was utilized to investigate the effect of asphalt mixture performance on moisture-induced damage. Also, to simulate the conditions of F-T cycles on the performance of asphalt mixtures under the influence of various forming minerals, ITST was used in F-T cycles of 1, 3, and 5. In order to perform the test by the modified Lattmann, three samples should be made in dry conditions and three in wet conditions. So, for the simulation of wet conditions, the specimens were first saturated with water for five minutes. They were then held for 5–10 minutes in drowned and without vacuum conditions, and then the saturated specimens were kept for 16 hours at −18°C in the freezer. They were then put in a water bath for 24 hours at temperature 60°C. The samples were finally brought to a temperature of 25°C and remained as is for 24 hours. In this experiment, loading was performed at a rate of 2 inches per minute till the specimens break. The amount of loading was filed at the failure moment. By the use of equation (8), the value of indirect tensile strength (ITS) of the samples was achieved [46, 47]:

\[
ITS = \frac{2F}{\pi n D}
\]

where \( F \) is the maximum load at the moment of failure (kN), \( t \) presents the thickness of the sample (m), and \( D \) presents the diameter of the sample (m). The moisture sensitivity of the sample with the average ITS ratio of the wet to dry samples was obtained from the following equation [48]:

\[
TSR = \frac{\text{ITS}_{\text{wet}}}{\text{ITS}_{\text{dry}}} \times 100,
\]

where ITS_{wet} represents the average ITS value in wet samples (kPa) and ITS_{dry} represents the average ITS value in dry samples (kPa).

4.5. Measuring the SFE Components of Bitumen and Aggregates. For calculating the aggregate SFE components, the universal sorption device (USD) was applied. In the USD method, it is possible to measure the surface components of aggregates in terms of their specific surface area (SSA) and then calculate the vapor distribution pressure of the three liquids on the aggregate surface. For each aggregate, there
are three SSA values, and the arithmetic mean of these is used. For the creation of a set of three unknowns (three SFE components of a solid material) and three equations, three relationships are needed to a probe liquid for each relationship. So to calculate the SFE components of solid material, three probe liquids, whose SFE components are already known, are needed [16, 22]. In this research, the probe liquids of n-hexane, methyl propyl ketone, and water were applied; Table 3 illustrates their SFE components.

For each liquid, a relation such as equation (10) must be formed.

\[ W_{V,S}^n = 2\Gamma_{v}^{total} + \pi_c = 2 \left( \sqrt{\Gamma_s^L \Gamma_w^L} + \sqrt{\Gamma_s^r \Gamma_w^r} + \sqrt{\Gamma_s^L \Gamma_w^r} \right) \]

(10)

where \( W_{V,S}^n \) is the adhesion work of vapor-aggregate surface, \( \Gamma_v^{total} \) is the total SFE of the vapor of a liquid, and \( \pi_c \) is the equilibrium distribution pressure corresponding to liquid-vapor on the surface of the aggregate that is achieved by its adsorption isotherm [49]:

\[ \pi_c = \frac{RT}{MA} \int_0^{p_0} \frac{n}{p} dp. \]

(11)

in which \( R \) presents the universal gas constant, \( M \) presents the molecular weight of a liquid-vapor, \( T \) represents the temperature of the test (Kelvin), \( A \) is the SSA of the aggregate, \( p_0 \) is the saturated vapor pressure, and \( n \) represents the mass of vapor absorbed per mass unit of the aggregate in the \( p \) vapor pressure. The \( S_v \) of aggregates is obtained through the classical equation of Brunauer, Emmett, and Teller (BET) as [49]

\[ A = \frac{n_m N_0}{M} \times \alpha, \]

(12)

where \( \alpha \) is the projected area of a molecule, \( n_m \) is the absorbed monolayer capacity on the aggregate surface, and \( N_0 \) is the Avogadro number. \( n_m \) is achieved by equation (13), in which \( S \) and \( I \) are the slope and intercept of the curve, respectively, which indicates \((p/(p_0 - p)n)\) versus \((p/p_0)\).

\[ n_m = \frac{1}{I + S}. \]

(13)

The contact angle between the bitumen and a liquid is achieved by the use of the Wilhelmy plate method, three bitumen SFE components are unknown with the \( s \) index, in that, if this test is repeated three times with various probe liquids, there will be three relationships in that there are three joint unknowns which were obtained by solving these three relationships simultaneously[49].

5. Results and Discussion

5.1. XRF Test Results. Different factors affect the occurrence of moisture damage, and the most important among them is the structure of the aggregate-forming minerals used in the asphalt mixture. Calcium oxide (CaO) and silicon dioxide (SiO2) minerals lead to a basic change in the hydrophilic or hydrophobic characteristics of the asphalt mixture. The higher the SiO2 content, the greater the hydrophilicity of aggregates, and vice versa. Conversely, the higher the percentage of the CaO mineral, the greater the hydrophobicity of the aggregates, and vice versa [20]. Therefore, the XRF test was performed to specify the structure of aggregate-forming minerals. According to this experiment, the compounds of the constituent minerals of the 8 aggregate types applied were calculated, and Figure 4 illustrates the results.

As is clear, most parts of the aggregates in mines 1–6 were composed of SiO2, which had strong acidic properties. In these types of aggregates, the amount of strong base parts, including CaO, was much less than the acidic part. Conversely, with respect to the aggregates of mines 7 and 8, it can be seen that in these aggregates, the percentage of the SiO2 mineral was very low compared to the CaO mineral, which can cause high adhesion resistance between aggregates and bitumen, which is due to the formation of insoluble bondings in water and the physical interaction between calcium on the surface of aggregates and bitumen. In addition to the minerals displayed in Figure 4, there were other minerals in aggregates that were very low in chemical analysis.

5.2. Mix Design Results. The optimal bitumen content of samples made with different forming minerals using the Marshall test is presented in Table 5. As it turns out, depending on the different forming minerals, the samples had a different content of optimal bitumen, although this difference was very small. However, the lower the amount of Al2O3 and SiO2 minerals and the higher the CaO mineral, the lower the optimal bitumen content in the aggregates corresponding to them.

5.3. Measuring the Bitumen Apparent Film Thickness. The apparent specific surface area (S_v) and bitumen apparent film thickness (AFT) on the surface of the aggregates used in this study based on equations (6) and (7) are represented in Table 6. As is clear, the bitumen coating on all aggregates was approximately 8 μm. In other words, since the bitumen used is the same for all aggregates, it can be said that the AFT on aggregates with different minerals does not differ.
5.4. ITST Results. According to the standards used, the numerical value of ITS alone cannot be used as an appropriate indicator to specify the moisture susceptibility of aggregates, but TSR is the most common indicator in specification of the moisture susceptibility of mixtures and predicting their performance. It should be noted that the

| Liquids | Acidic (Γ⁺) | Basic (Γ⁻) | Polar (Γ⁴B) | Nonpolar (Γ⁴LW) | Total SFE, (Γ) |
|---------|-------------|-------------|-------------|-----------------|---------------|
| n-Hexane | 0           | 0           | 0           | 18.4            | 18.4          |
| Methyl propyl ketone | 0 | 19.6        | 0           | 24.7            | 24.7          |
| Water   | 25.5        | 25.5        | 51          | 21.8            | 72.8          |

Table 4: SFE components of materials in order to calculate the same components for bitumens.

| Liquids | Acidic (Γ⁺) | Basic (Γ⁻) | Polar (Γ⁴B) | Nonpolar (Γ⁴LW) | Total SFE, (Γ) |
|---------|-------------|-------------|-------------|-----------------|---------------|
| Formamide | 2.28        | 39.6        | 19          | 39              | 58            |
| Water   | 25.5        | 25.5        | 51          | 21.6            | 72.6          |
| Glycerol | 3.92        | 57.4        | 28.8        | 34              | 62.8          |

Table 5: Optimal bitumen content of samples made with different aggregates.

| Mines | Bitumen content corresponding to Marshall maximum strength (%) | Bitumen content corresponding to the maximum specific gravity of the mixture (%) | Bitumen content corresponding to 4% air voids (%) | Optimal bitumen content (%) |
|-------|-------------------------------------------------------------|---------------------------------------------------------------|-------------------------------------------------|------------------------------|
| 1     | 5.7                                                         | 5.7                                                          | 6.5                                             | 5.9                          |
| 2     | 5.7                                                         | 5.7                                                          | 6.5                                             | 5.9                          |
| 3     | 5.7                                                         | 5.7                                                          | 6                                               | 5.8                          |
| 4     | 5.7                                                         | 5.6                                                          | 6                                               | 5.8                          |
| 5     | 5.7                                                         | 5.7                                                          | 6                                               | 5.8                          |
| 6     | 5.7                                                         | 5.6                                                          | 5.8                                             | 5.7                          |
| 7     | 5.6                                                         | 5.5                                                          | 5.7                                             | 5.6                          |
| 8     | 5.4                                                         | 5.3                                                          | 5.6                                             | 5.4                          |

Table 6: Measuring the bitumen apparent film thickness.

| Mines | P_b (%) | P_t (%) | G_b (kg/m³) | G_t (kg/m³) | G_mm (kg/m³) | V_b (%) | V_t (%) | V_mm (%) | V_a (%) | S_a (m²/kg) | AFT (μm) |
|-------|---------|---------|-------------|-------------|--------------|---------|---------|-----------|---------|-------------|---------|
| 1     | 5.90    | 94.12   | 2.53        | 1.02        | 2.44         | 2.36    | 3.20    | 14.07     | 4.62    | 9.44        | 5.31    | 8.004    |
| 2     | 5.90    | 94.05   | 2.53        | 1.02        | 2.44         | 2.36    | 3.32    | 14.23     | 4.80    | 9.43        | 5.31    | 8.001    |
| 3     | 5.80    | 94.66   | 2.53        | 1.02        | 2.43         | 2.32    | 4.53    | 12.72     | 3.40    | 9.32        | 5.31    | 7.992    |
| 4     | 5.80    | 94.12   | 2.53        | 1.02        | 2.44         | 2.36    | 3.20    | 14.07     | 4.62    | 9.44        | 5.31    | 8.004    |
| 5     | 5.80    | 94.23   | 2.55        | 1.02        | 2.46         | 2.37    | 3.78    | 13.92     | 4.53    | 9.39        | 5.26    | 7.994    |
| 6     | 5.70    | 94.54   | 2.57        | 1.02        | 2.47         | 2.35    | 4.90    | 13.22     | 3.93    | 9.29        | 5.23    | 7.995    |
| 7     | 5.60    | 94.15   | 2.54        | 1.02        | 2.45         | 2.35    | 4.16    | 14.05     | 4.69    | 9.36        | 5.29    | 7.997    |
| 8     | 5.40    | 94.55   | 2.55        | 1.02        | 2.45         | 2.36    | 3.80    | 13.09     | 3.71    | 9.38        | 5.26    | 7.992    |
TSR values are always less than 100%, because by placing the samples in F-T cycles, ITS of the samples decreases compared to the dry samples [51]. The results of ITS and TSR of samples with different aggregates under F-T cycles of 1, 3, and 5 are shown in Figures 5 and 6, respectively. As can be seen, samples constructed with aggregates of mines 7 and 8 (with high CaO mineral content) experienced a higher tensile resistance than other samples in all F-T cycles. Moreover, TSR amounts of samples decreased with increasing F-T cycles. It is also observed that aggregates with greater CaO and less SiO2 amounts had the greatest resistance to moisture-induced damage, and aggregates with lower CaO and higher SiO2 amounts had the lowest resistance to moisture-induced damage. Moreover, samples made with mine aggregates of 1–8, in the first cycle, had a 47.7%, 43.3%, 39.6%, 35.8%, 32.3%, 30.5%, 25.8%, and 21.6% decrease in TSR, respectively, compared to the dry samples. Also, in the third F-T cycle, they experienced a 61.9%, 57.6%, 54.2%, 51.8%, 49.7%, 48.6%, 45.6%, and 42.5% TSR drop, respectively, compared to the dry samples, and in the fifth cycle, they had 79.6%, 78.1%, 76.8%, 75.2%, 74.1%, 72.9%, 71.7%, and 69.6%, respectively, drop in TSR value. Thus, the samples constructed with the aggregates of mine 8 indicated the lowest decrease (44.56%, on average) in moisture resistance during F-T cycles.

Numerous factors are effective in the resistance of asphalt mixtures to moisture-induced damage, one of the main causes of which is the structure of the minerals forming the aggregates of an asphalt mixture. In other words, the presence of minerals such as SiO2 and CaO can have a great impact on the hydrophilic and hydrophobic properties of aggregates. Therefore, according to the obtained results, it is clear that the aggregates of mines 1–6 had the highest SiO2 mineral value, while the CaO mineral value in the aggregates of mines 7 and 8 was higher, which is evidence of their hydrophilicity and hydrophobicity, respectively. Of course, not only the type of minerals that make up aggregates affect the resistance of mixtures to moisture but also the surface texture of aggregates, the specific surface area, the type of bitumen used, and its compatibility with aggregates, additives, and fillers also affect the resistance of mixtures against moisture.

5.5. SFE Test Results. The results of calculating the SFE components of aggregates with different minerals are presented in Table 7. As is clear, aggregates with more calcareous properties had a larger basic component and a smaller acidic component, compared to other aggregates. The large basic component in these types of aggregates makes stronger bondings between these groups of aggregates and bitumen, which has acidic characteristics, and stripping is less likely to be expected. Also, in mines where the nonpolar component of their aggregates was larger, the tendency of the aggregates to adherence and wettability by water decreased, and this increases the adhesion of aggregates to bitumen, which is a nonpolar substance, by forming covalent bondings. In addition, increasing the nonpolar characteristics indicates a greater inclination of aggregates to construct nonpolar bonds and their greater resistance to damage caused by moisture. Also, the total SFE of aggregates of mines 7 and 8 was more than the others.

Table 8 presents the results of calculating the bitumen SFE components. As is clear, the bitumen acidic component was remarkably larger than its basic component, indicating that bitumen has acidic characteristics. Also, a large part of bitumen SFE was its nonpolar component. As a matter of fact, most of the adhesion formed between aggregate and bitumen is due to nonpolar or covalent bondings.

The results of adhesion SFE between the 8 types of aggregates and bitumen are given in Table 9. The column results of AFE of bitumen aggregate showed that AFE between bitumen and aggregates with higher CaO and less SiO2 amounts was more than AFE between bitumen and aggregates with lower CaO and higher SiO2 amounts, indicating that in dry conditions (without the presence of water), it is more difficult to detach the bitumen from the surface unit of aggregates with more calcareous properties than other aggregates and needs further energy. In fact, the presence of more CaO mineral and less SiO2 mineral caused AFE to increase, and by increasing CaO amount and reducing the SiO2 amount, the energy required for rupture at the bitumen-aggregate contact surface increased in dry conditions. Also, according to the construction of samples only with 60–70 penetration grade bitumen, AFE of asphalt-water for all 8 types of aggregates was 57.2 erg/cm².

In all specimens, the AFE of the bitumen-aggregate system changed from a positive amount in dry conditions to a negative amount in moisture. Therefore, when the three materials of bitumen, water, and aggregate come in contact, water alters the system free energy to reach the lowest energy state so that the phenomenon of stripping occurs. Samples made with aggregates with less calcareous properties had a lower negative AFE in the presence of water than aggregates with more calcareous properties. In other words, as the bitumen separates from the surface unit of the aggregate by water infiltration, further energy is unleashed, and therefore the tendency is greater for stripping in these aggregates. The presence of higher amounts of CaO in aggregates increased AFE of bitumen-aggregate in the presence of water to zero, indicating that the desire of the system for stripping and obtaining a stable level in which its energy level is the least is decreased. An increase in the CaO amount and a decrease in the SiO2 amount reduce this tendency.

5.6. The Effect of Aggregates-Forming Minerals on Moisture Susceptibility and SFE. In this section, the results of the effect of different minerals of aggregates on the moisture susceptibility of asphalt mixtures in 1, 3, and 5 F-T cycles, and SFE of aggregates were investigated using the linear regression analysis.

5.6.1. The Effect of CaO and SiO2 on Moisture Susceptibility. Table 10 shows the coefficients of linear regression analysis, along with the significance tests for TSR after 1, 3, and 5 F-T cycles. The coefficients of the regression model are the same as the Beta coefficient column. Any variable with a higher
Figure 5: ITST results in samples made with various forming minerals in different F-T cycles.

Figure 6: TSR values for samples made from aggregates of different mines.

Table 7: SFE components of aggregates used (ergs/cm²).

| Mines | Acidic ($\Gamma^+$) | Basic ($\Gamma^-$) | Polar ($\Gamma^{AB}$) | Nonpolar ($\Gamma^{LW}$) | Total SFE ($\Gamma$) |
|-------|---------------------|--------------------|-----------------------|--------------------------|----------------------|
| 1     | 24.8                | 36.9               | 60.5                  | 22.6                     | 83.1                 |
| 2     | 22.0                | 64.6               | 75.4                  | 26.9                     | 102.2                |
| 3     | 19.6                | 81.4               | 79.9                  | 29.6                     | 109.5                |
| 4     | 18.4                | 163.5              | 109.7                 | 33.9                     | 143.6                |
| 5     | 15.1                | 216.9              | 114.5                 | 40.7                     | 155.2                |
| 6     | 13.7                | 326.5              | 133.8                 | 49.1                     | 182.9                |
| 7     | 12.2                | 401.1              | 139.9                 | 52.0                     | 191.9                |
| 8     | 10.9                | 446.7              | 139.6                 | 52.9                     | 192.5                |
Beta coefficient is more important in the regression model. Given the smaller \( p \) value of each variable than 0.05, their null hypothesis is rejected [52–54]. Based on the previously obtained results, CaO and SiO\(_2\) minerals had the greatest impact on moisture susceptibility and also formed a large part of aggregates according to the XRF test in these 8 mines. Also, considering all the minerals in regression, many minerals, except the important minerals CaO and SiO\(_2\), were removed from the analysis due to the collinearity. Therefore, it was decided to consider only CaO and SiO\(_2\) minerals in the analysis of moisture susceptibility. Therefore, the effect of these two minerals on different cycles was measured. The results showed that due to the positive regression coefficient of the CaO mineral, it had a positive effect on TSR, and on the other hand, due to the negative regression coefficient of the SiO\(_2\) mineral, it had a negative impact on the TSR of mixtures. Also, as it is clear, in cycles 1, 3, and 5, the regression coefficient value of CaO was 1.045, 0.713, and 0.339, respectively, which indicates that the content change of the CaO mineral, a more obvious influence on TSR was observed in cycle 1. The results of the SiO\(_2\) mineral also showed that for a unit change in this variable, a negative effect on TSR was observed. Also, according to the data in the Table, it can be said that all variables had significant coefficients on moisture sensitivity of asphalt mixtures [55, 56].

5.6.2. The Effect of CaO and SiO\(_2\) on SFE. Table 11 illustrates the regression analysis results of the SFE components. The results of the minerals on the SFE acidic component show that the CaO mineral had a negative effect on this component due to the negative Beta coefficient \((-0.463)\) while the SiO\(_2\) mineral had a positive effect on it \(0.713\). The positive regression coefficients of CaO mineral in basic, polar, and nonpolar components and total SFE showed a positive effect of this mineral on the aforementioned components and total SFE, in which the regression coefficient values of CaO mineral were 3.252, 2.947, 6.055, and 6.268, respectively. On the other hand, the negative regression coefficient of the SiO\(_2\) mineral on these components and the total SFE showed a negative effect of this mineral on these components and the total SFE and the regression coefficient value of the SiO\(_2\) mineral were \(-2.489, -2.912, -2.965, \) and \(-4.478\), respectively.

| Mines | Type of bitumen | AFE values |
|-------|-----------------|------------|
| 1     | 60.5            | −94.8      |
| 2     | 68.7            | −103.5     |
| 3     | 72.8            | −88.6      |
| 4     | 85.8            | −64.5      |
| 5     | 94.7            | −52.6      |
| 6     | 108.2           | −25.4      |
| 7     | 116.1           | −19.9      |
| 8     | 115.4           | −7.1       |

| Dependent variables | Independent variables | Unstandardized coefficients | Standardized coefficients |
|---------------------|-----------------------|----------------------------|--------------------------|
|                     |                       | B  | Std. error | Beta | t  | Sig. |
| Cycle 1             | CaO                   | 1.045 | 0.103    | 0.428 | 10.134 | 0.010 |
|                     | SiO\(_2\)             | −0.546 | 0.059   | −0.397 | −9.216 | 0.012 |
| Cycle 3             | CaO                   | 0.713 | 0.090    | 0.400 | 7.941 | 0.015 |
|                     | SiO\(_2\)             | −0.463 | 0.052   | −0.460 | −8.973 | 0.012 |
| Cycle 5             | CaO                   | 0.339 | 0.056    | 0.358 | 6.492 | 0.026 |
|                     | SiO\(_2\)             | −0.257 | 0.032   | −0.480 | −8.015 | 0.015 |

| Mines | Type of bitumen | SFE components | Bitumen-aggregate | Asphalt-water | Bitumen-aggregate in the presence of water |
|-------|-----------------|----------------|------------------|---------------|------------------------------------------|
| 60–70 | 1.8             | 0.68           | 2.27             | 14.01         | 16.28                                    |

**Table 9: Adhesion free energy values (erg/cm\(^2\)).**

**Table 10: Analysis of the effect of minerals on moisture susceptibility after 1, 3, and 5 F-T cycles.**

**Table 8: Bitumen SFE components (ergs/cm\(^2\)).**

**Table 11: Bitumen SFE components (ergs/cm\(^2\)).**

**Table 12: Adhesion free energy values (erg/cm\(^2\)).**

**Table 13: Analysis of the effect of minerals on moisture susceptibility after 1, 3, and 5 F-T cycles.**
Results showed that the CaO mineral had the highest effect on total SFE positively, and SiO₂ mineral had the most significant effect on total SFE results negatively.

6. Conclusion

One of the important parameters in determining the moisture susceptibility of different aggregates is the structure of aggregate minerals. The relationship between aggregate-forming minerals and surface free energy can also help select more resistant aggregates. Therefore, in this study, by examining 8 types of aggregates with different minerals, the constituent structure of each of them on the moisture sensitivity of asphalt mixtures was explored. The most significant results of this study are:

(i) The XRF test was done to specify the structure of aggregate-forming minerals, and it was indicated that most parts of the aggregates in mines 1–6 were composed of SiO₂, which had strong acidic properties, according to acidic components of SFE results. In fact, the percentage of the SiO₂ mineral in the aggregates of mines 7 and 8 was very low compared to the CaO mineral, which can cause high adhesion resistance between aggregates and bitumen. Also, other minerals in the aggregates had very low amounts.

(ii) The results of measuring the bitumen apparent film thickness showed that the bitumen coating on all aggregates was approximately 8 μm.

(iii) ITST results indicated that samples constructed with aggregates of mines 7 and 8 (with high CaO mineral content) experienced a greater tensile resistance than other samples in all F-T cycles. Moreover, TSR amounts of samples reduced with increasing F-T cycles. Also, aggregates with higher CaO and less SiO₂ amounts had the greatest resistance against moisture-induced damage.

(iv) The samples made with the aggregates of mine 8 showed the lowest decrease (44.56%, on average) in TSR during F-T cycles.

(v) Results indicated that the structure of aggregate-forming minerals was effective in the resistance of asphalt mixtures to moisture-induced damage. In other words, the presence of minerals such as SiO₂ and CaO had a great impact on the hydrophilic and hydrophobic properties of aggregates, respectively, in ITST and SFE tests.

(vi) Results of the SFE method showed that aggregates with more calcareous properties had a smaller acidic component and a larger basic component compared to other aggregates, which makes stronger bonds between these groups of aggregates and bitumen, and stripping is less likely to be expected.

(vii) In mines 7 and 8 where the nonpolar component of their aggregates was larger, the tendency of the aggregates to adherence and wettability by water decreased and this increases the adhesion of aggregates to bitumen and their resistance to damage caused by moisture.

(viii) The SFE results also showed that the total SFE of aggregates of mines 7 and 8 was more than the others.

(ix) The results of calculating the bitumen SFE components indicated that the bitumen acidic component was remarkably larger than its basic component. In addition, a large part of bitumen SFE was its nonpolar component. As a matter of fact, most of the adhesion formed between aggregate and bitumen is due to nonpolar or covalent bondings.

(x) The SFE method results showed that the AFE between bitumen and aggregates with higher CaO and lesser SiO₂ amounts was more than the AFE between bitumen and aggregates with lower CaO and higher SiO₂ amounts, indicating that in dry conditions, more energy is required to detach the bitumen from the surface unit of aggregates with more calcareous characteristics.

(xi) The presence of greater amounts of the CaO mineral in aggregates increased the AFE of bitumen-aggregate in the presence of water to zero, illustrating that the desire of the system to the
phenomenon of stripping is decreased. An increase in the CaO amount and a decrease in the SiO₂ amount decreased this tendency.

(xii) The effect of mineral type on moisture susceptibility by linear regression analysis showed that due to the positive regression coefficient of the CaO mineral, it had a positive impact on TSR; on the other hand, due to the negative regression coefficient of the SiO₂ mineral, it had a negative effect on the TSR of mixtures.

(xiii) The effect of mineral type on the SFE also indicated that the CaO had a negative effect on acidic component due to the negative Beta coefficient, while the SiO₂ had a positive effect on it.

(xiv) The positive regression coefficient of CaO in basic, polar, and nonpolar components, and the total SFE illustrated a positive effect of this mineral on these components and total SFE, and the negative regression coefficients of the SiO₂ showed a negative effect of this mineral on these components and the total SFE.

Data Availability

The data used to support the findings of this study are currently under embargo, while the research findings are commercialized. Requests for data, 3 months after the publication of this article, will be considered by the corresponding author.

Conflicts of Interest

The authors declare that they have no conflicts of interest reported in this paper.

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