**Evaluating the cracking resistance of Hot Mix Asphalt modified with calcium lignosulfonate at low and intermediate temperatures**

Seyed Ali Ziaee[*1], Seyed Saeed Fatemi[2], Jafar Bolouri Bazaz[3], Rouzbeh Shad[4], Seyed Amir Saadatjoo[5]

[*1]Department of Civil Engineering, Faculty of Engineering, Ferdowsi University of Mashhad, Mashhad, Iran. Email: sa-ziaee@um.ac.ir (corresponding author), Mobile Number: +989155128094

[2]Department of Civil Engineering, Faculty of Engineering, Ferdowsi University of Mashhad, Mashhad, Iran. Email: sfomran@mail.um.ac.ir, Mobile Number: +989152007750

[3]Department of Civil Engineering, Faculty of Engineering, Ferdowsi University of Mashhad, Mashhad, Iran. Email: bolouri@um.ac.ir, Mobile Number: +989155128094

[4]Department of Civil Engineering, Faculty of Engineering, Ferdowsi University of Mashhad, Mashhad, Iran. Email: r.shad@um.ac.ir, Mobile Number: +0989155128094

[5]Department of Civil Engineering, Iran University of Science and Technology, Tehran, Iran. Email: amir.saadatjoo@gmail.com, Mobile Number: +989151045127

**Abstract**

One of the common types of failures in asphalt pavements is cracking. Considering the environmental importance of material recycling, in this research, calcium lignosulfonate (CLS) as a waste product of cellulose production, was used as a bitumen additive, and the cracking resistance of semi-circular bending (SCB) samples of asphalt mixtures containing bitumen modified with 5% to 20% CLS was investigated at low and medium temperatures. To measure the performance of asphalt mixtures, the parameters of fracture energy ($G_f$), fracture toughness ($K_{1c}$), flexibility index (FI) and cracking resistance index (CRI) were used. Also, rotational viscosity (RV) test was performed to determine the pumpability of bitumens. The results showed that CLS particles can increase the stiffness and viscosity of bitumen in high-temperature conditions. However, it was lower than the maximum allowed amount. At low temperature, with the addition of CLS, $K_{1c}$ and $G_f$ indices were increased, and their highest values were related to bitumen with a 15% additive. At intermediate temperature, $G_f$ index was increased until before the critical load, and FI and CRI were increased up to 15% and then decreased slightly. But $G_f$ index was increased with a decreasing trend after the critical load until the end.

**Keywords:** Asphalt mixture, recycled bituminous additive, fracture toughness, fracture energy, flexibility index, cracking resistance index

## 1. Introduction

One of the common types of failures in asphalt pavements is cracking. Asphalt mixture resistance versus cracking is one of the most important parameters that are investigated for improving the mixtures' performances. Asphalt mixture cracking mostly happens at a low temperature because of asphalt mixtures' brittle behaviour which causes a high cost for renovation organizations. These kinds of cracking, which are called low-temperature crack (LTC), have a remarkable influence on the weakness of pavement structures [1-3]. Moreover, based on the viscoelastic behaviour of asphalts, an intermediate temperature crack (ITC) has also been presented as one of the serious cracking in an asphalt mixture [4-6].

Asphalt cracking has different factors, divided into primary as well as secondary ones. Primary factors include environmental factors and those related to the type of materials. Environmental factors are associated with stresses due to the temperature change in an inner layer of asphalts [7]. Factors related to the type of materials are also discussed in connection with the adhesion of bitumen and aggregates [8]. The secondary factors are related to pavement loading and traffic of vehicles on its surface, especially the traffic of heavy vehicles on the surface of the pavement which causes the deterioration of failures and cracks on its surface [9-13]. In general, it can be said that environmental factors and types of materials cause cracks, and factors related to pavement loading result in crack growth and expansion [14].

Cracks can be classified into fatigue cracks, reflection cracks (from top to bottom) and top to bottom cracks [15]. Cracks from top to bottom are longitudinal and transverse cracks that start in the surface layers of the asphalt pavement and extend downwards. Various reasons cause cracks from top to bottom, which can be mentioned as the stresses created in the contact surface of vehicle tires with the pavement, stresses due to the temperature change of asphalt pavement and aging of the asphalts [16].

Due to the fact that the asphalt mixture is a viscoelastic material [17], its fracture resistance characteristic depends on temperature, time and loading rate [18]. For this purpose, a detailed assessment of mechanical features of asphalt mixtures fracture and the factors affecting it to bring the laboratory conditions closer to the real and local conditions will be one of the effective measures in identifying and improving the pavement [19].

Many fracture tests have been used in order to investigate the fracture behaviour of asphalts, among which Edge notched disc bend (ENDB), Semi-circular Bending (SCB), Double-edged Notched Tension (DENT), Disc-shaped Compact Tension (DCT) and Single-edge Notched Beam (SENB) test and can be mentioned [20-26]. Among the different methods of fracture tests, SCB test has been used further than other samples, the main reason for which is the ease of making and cutting the samples, as well as being close to the field conditions [4, 7, 14, 27-29]. This test was used for the first time to study the fracture resistance of rock material samples [30]. Also, these samples have been used to investigate the cracking resistance of asphalts at the low temperature [31-33] and intermediate temperature [34-36].

As mentioned earlier, cracking is caused by various causes, including weakness in the properties of asphalt materials, such as aggregates and bitumen. Therefore, any improvement in the properties of these two components will change the final properties of the mixture. Considering the mixing and dispersion of asphalt binder in the whole specimen and its importance in the adhesion of aggregates and asphalt cracking resistance, many researchers has investigated the influence of bitumen additives on the performance of asphalts, some of these materials can be classified into one of the general classifications including polymer-based additives, nanomaterials, fibers and recycled and biological materials [34, 37-39]. Recycled and biological additives have received more attention in recent years. Because they improve the properties of asphalts and prevent the accumulation of waste from various industries and help to preserve the environment. In this context, various studies have been conducted on the evaluation of the influence of crumb rubber on the behavior of asphalts and have shown that using this material represents positive effects on the cracking as well as rutting resistances of asphalts [40-43].

Lignin is one of the biomaterials that has been considered for enhancing asphalts' performance. Lignin is one of the most abundant natural polymers, which is composed of phenylpropane units and is mainly obtained from timber and pulp production [44, 45]. As an industrial waste material, more than 50 million tons of it are produced annually. But only half of it is consumed and the rest is stored untouched [46, 47]. Thus, one of the main concerns of researchers is to find an approach for reusing this substance in industries. For this purpose, this substance has been used in some studies as a modifier in concrete as well as asphalt mixtures, and it has been illustrated that using this substance in the pavement enhances various performance characteristics of bitumen and asphalt mixture. Moreover, it presents environmental advantages [44, 48]. Also, investigators stated that though adding wood lignin into asphalt binder positively influences the rutting behavior of specimen, increasing the asphalt binder's viscosity as well as mixture's brittleness has a straightforward relationship with lignin amounts [49]. Calcium lignosulfonates (CLSs) are the amorphous brown powder extracted from sulfite pulp in cellulose productions [46, 50]. It has hydrophobic and hydrophilic groups, so the solubility potential of CLS in an asphalt binder is higher compared to that of lignin [51]. In addition, it is susceptible to be used as an anti-aging material for bitumen since the chemical structures of CLS have a similarity with those of petroleum bitumens [52]. In this regard, some studies have investigated the effect of this material on asphalt mixtures' performances.

In the initial research that was conducted on the properties of an asphalt mixture comprising CLS, the improvement of the rutting behavior of asphalt sample was seen as a result of using percentages of 5 to 10 percent. In the subsequent research, it was found that the use of this material, along with the improvement of rutting, increases the viscosity of the bitumen and the brittleness of the mixture [49]. Batista et al. used lignin kraft for bitumen modification of asphalt mixtures. In this research, samples modified with this material showed higher rutting resistance than unmodified samples. Also, the examination of low temperatures showed that the use of this material increases the toughness at low temperatures [53]. While in McCready and Williams's research, the increase in rutting behavior at high temperatures was confirmed, but the behavior at low temperature showed a slight decrease [54]. Fatemi et al. investigated the effect of adding CLS at 5, 10, 15 and 20 percent. In this study, it was found that all percentages of CLS have anti-aging properties and also 15% value provides the greatest resistance to creasing [55-57]. In several separate researches, Zarei et al. investigated various properties of CLS, including cracking resistance, moisture sensitivity in different thaw and freeze cycles, along with some materials such as fibers. The results of these researches showed that CLS can improve the fracture toughness in different loading modes as well as different temperatures. These researchers also pointed out that CLS can be considered as a substance that increases the cracking resistance of the mixture. In this collection of researches, it is stated that CLS can improve stripping by increasing toughness [58-61]. According to the studies that have been done so far regarding the influence of this material on the performance of asphalt samples and considering the importance of the phenomenon of cracking in asphalt, the novelty of this article is that the influence on the fracture behaviour of asphalt samples at low and intermediate temperatures has been investigated simultaneously with rotational viscosity (RV) test to determine the optimum percentage of CLS additive to have an asphalt mixtures with reasonable performance and bitumens with feasible workability.

## 2. Objective

In the current article, the cracking resistance of asphalt samples comprising bitumen modified with calcium lignosulfonate has been investigated at low and intermediate temperatures. For this purpose, bitumens modified with 5, 10, 15 and 20% by weight of calcium lignosulfonate bitumen were used to make warm mix asphalt. Then, rotational viscosity (RV) test was performed on different bitumens to determine the pumpability and feasibility of workability of bitumen containing different percentages of additive. Next, by making semi-circular bending samples, the parameters of fracture energy ($G_f$) as well as fracture toughness ($K_{1c}$) were calculated for negative temperature samples, and parameters of $G_f$ and flexibility index (FI) and cracking resistance index (CRI) were calculated for intermediate temperature samples. Finally, the values of the mentioned indices for mixtures with different bitumens were compared with each other in order to determine the appropriate amount of additive for the best performance of the asphalt mixture modified with calcium lignosulfonate, taking into account its workability.

## 3. Materials and construction of samples

### 3.1. Aggregates

In this article, the aggregates and fillers used in one of the main aggregate production mines in the northeast of the country around Mashhad city have been prepared. The aggregates' physical characteristics are represented by Table 1 and compared with their standard values that are within the allowed range.

Considering that in Iran, most of the roads are designed and implemented based on grading No. 4 of the Iranian Road Pavement Regulations [62], the grading of aggregates used in this article is selected the same grading. The upper and lower limits and the selected grading are shown in Figure 1.

### 3.2. Bitumen

Due to the widespread use of bitumen with 60-70 penetration grade in road construction in Iran [63], the same bitumen was used in this research, which was obtained from Naft Sharq in Mashhad. Table 2 shows the physical characteristics of bitumen used in this research.

### 3.3. Calcium lignosulfonate

In this research, calcium lignosulfonate additive was used as a solution in bitumen for improving the characteristics of asphalt mixtures. This additive was provided from Ligno Tech South Africa. Table 3 shows the chemical compounds in the calcium lignosulfonate material used in this research. Table 4 also shows the physical characteristics of this additive. A calcium lignosulfonate powder additive can also be seen in Figure 2.

### 3.4. Mix design and making samples

For the preparation of asphalt mixture, firstly, aggregate grading was selected according to the middle range of grading curves of No. 4 of Iran's Road Pavment Regulations and Report 234 for the Topeka layer. The next step in making the samples is to determine the optimal bitumen percentage based on the ASTM D1559 standard and provide the asphalt mixture mix design. Based on Marshall mix design, asphalt samples were prepared with 75 blows of Marshall hammer and with six various bitumen amounts of 4, 4.5, 5, 5.5, 6 and 6.5% and with three repetitions of each sample. After making asphalt mixtures using the Marshall machine, each of the samples was subjected to the Marshall stability test as well as Marshall number and flow value of each sample were obtained. Afterwards, the optimal value of bitumen for mix design was determined based on the average of three amounts of bitumen corresponding to the highest Marshall stability, the amount of bitumen corresponding to the highest specific gravity, and the amount of bitumen corresponding to the air void percentage of 4%. After calculations, the optimal bitumen percentage of asphalt mixtures was found to be 4.9%, and this value was considered the same for making all asphalt mixtures, both control and modified ones.

To mix CLS in pure bitumen, based on the experiences of previous studies, the bitumen was heated to 160°C and then added CLS to it and continued mixing for 30 minutes using a high-shear mixer at 3000 rpm [64]. The results of the research carried out by Jedrzejczal et al. showed that the use of more than 20% CLS to modify bitumen properties is not economically viable [65]. Therefore, in this research, CLS additive with 5, 10, 15 and 20 percent by binder weight was used for bitumen modification and mixing in it. In Figure 3, a sample of bitumen is shown in additive mixing.

On the other hand, the aggregates were placed at 170 degrees for 18 hours and after the bitumens were prepared, the aggregates and pure or modified bitumen with 5, 10, 15 and 20% additives were mixed with each other and 2 samples of each combination were produced.

The prepared mixtures were poured into the Superpave gyratory compactor for compaction with the conditions listed in Table 5. Then the manufactured gyratory samples were cut to make the desired SCB samples. For this purpose, first, 2 cm was cut from the top and bottom of specimen. Then, 3 rings with a thickness of 27 mm were cut from a sample of each polymer percentage for negative temperature tests, and after cutting in half, a 20 mm groove was created in the middle. Also, 30 mm rings were cut from another sample of each percentage for positive temperature tests, and after cutting in half, a 15 mm groove was also created in the middle of them. Table 6 shows ID, polymer percentage and thickness of each sample.

## 4. Laboratory program

The experiments conducted in this research are divided into three general categories. The first group of tests related to the rotational viscosity test on different bitumens was to determine the pumpability and feasibility of workability of bitumen containing different percentages of additives. The second group of tests were conducted at -15 degrees to measure the low-temperature cracking resistance of the mixture with the use of fracture energy as well as toughness parameters, and the third group of tests that were conducted at +15 degrees for measuring the cracking resistance of mixture with the use of fracture energy, flexibility index and cracking resistance index parameters.

### 4.1. Rotational viscosity test (RV)

The viscosity test is performed with the aim of checking the pumpability and feasibility of workability of asphalt mixture during implementation [66]. In this test, according to the method provided in ASTM D4402, a cylindrical spindle that has a fixed rotation speed should be immersed in the binder [67]. In the current research, RV experiment was performed by Brookfield rotary viscometer with 20 rpm rotation speed at 135 degrees to evaluate the performance of bitumen in control and modified samples [68]. ASTM D4402 suggests the maximum viscosity value at 135 °C equal to 3 pascal.seconds and declares that its higher value indicates the impermissible viscosity of the bitumen and the lack of proper workability in the asphalt mixture during pavement implementation [69]. The rotational viscosity measuring device is shown in Figure 4. Also, bitumen samples made for RV testing are presented in Table 7.

### 4.2. Low-temperature cracking resistance test

To measure the cracking resistance of SCB samples of a mixture at -15 degrees, first these samples are kept for 8 hours at the target temperature until all the components of the sample reach that temperature. Then, the samples as shown in Figure 5 are subjected to monotonic (static) loading at a speed of 3 mm/min with the Universal Testing Machine (UTM) device. The way the sample is placed under the device is in such a way that the crack is completely aligned with the loading jaw and the distance between the supports is 12 cm. As a result, loading is applied to mode I and as pure tension. Cracking resistance indices at negative temperature where asphalt exhibits elastic behavior [70, 71], fracture energy ($G_f$) and fracture toughness ($K_{1c}$) are considered. The fracture energy parameter is obtained from the surface under the force-displacement curve, and the fracture toughness parameter for samples with 27 mm thickness and 20 mm groove is calculated as follows:

$$K_{1c} = \frac{P_{cr}}{2Rt} Y_1 \sqrt{\pi a} \qquad (1)$$

where a denotes the depth of the created groove in meters, R and t represent respectively the radius as well as thickness of sample in meters, $P_{cr}$ is the maximum tolerable load of sample in kN, and Y is the shape coefficient of the sample obtained by Abaqus [72].

The fracture energy calculated in this article is measured as the fracture energy before the critical load and fracture energy after critical load, which are respectively known as the stored fracture energy and the residual fracture energy [73-76] and is obtained from the calculation of the area under the force-displacement curve up to the peak point of load and after the peak point of load, which is shown in Figure 6. The stored fracture energy indicates the resistance to cracking and the residual fracture energy indicates the resistance to crack propagation [73, 74, 77].

In general, the fracture energy achieved from the calculation of the area under the force-displacement diagram can be achieved by dividing the area under the force-displacement curve by the cracked cross-sectional area.

$$G_F = \frac{W_f}{A} = \int (P)\, du / A \qquad (2)$$

In which:

U = displacement in the direction of load application

P = force or load (KN)

$W_F$ = work due to failure

$G_F$ = final fracture energy ($J/m^2$)

A = crack-free surface (equal to the product of the crack-free line and the thickness of the sample)

**4.3. Intermediate temperature cracking resistance test**

SCB samples and monotonic loading were used to measure cracking resistance at intermediate temperature like low temperature. With the difference that the thickness of the samples was 30 mm, the groove depth was 15 mm and the loading speed was 0.3 mm/min. The other loading conditions were considered the same as the low temperature condition. In this way, the maximum load that can be tolerated by the sample was calculated by the UTM device and the force-displacement diagram of each sample was extracted as the output of the device. According to the explanations provided in section 4.2, the fracture energy of the samples at intermediate temperature was considered as the fracture energy before the critical load and the fracture energy after the critical load.

One of the parameters that was calculated at intermediate temperature is the flexibility index (FI). FI is a parameter to identify brittle mixtures that are sensitive to premature cracks. The studies of

Al-Qadi et al. [78] showed that the energy required to cause fracture in the sample at intermediate temperature alone is not a suitable indicator for measuring cracking resistance, so this index was introduced and used for the first time in 2016 at the University of Illinois, USA, for evaluating mixture's resistance against cracking at intermediate temperature, which is obtained from the following relationship:

$$FI = \frac{G_f}{|m|} \times 0.01 \quad (3)$$

In which:

$G_F$ = final fracture energy (J/m$^2$)

m = slope at the inflection point of second part of force-displacement curve in kN/mm

In SCB samples of asphalt mixture, the higher the FI index, it means that the sample shows more resistance to crack propagation [79].

In 2018, Kaseer et al. [80] developed an index for evaluating brittle cracking potentials of a mixture by SCB samples. In the current research, a cracking parameter, known as the cracking resistance index (CRI), was obtained from the response of the load-displacement curve while the testing of SCB samples, and its comparison was made with the developed flexibility index (FI). As it mentioned, at low temperature asphalt has brittle behavior. So there is no slope in the second part of force-displacement curve and CRI can be calculated instead.

The CRI index is calculated as follows:

$$CRI = \frac{G_f}{|P_{max}|} \quad (4)$$

In this relation, $G_f$ is equal to the total amount of energy required to break the asphalt mixture in J/m$^2$ and $P_{mas}$ is the maximum force applied to the sample in terms of kN. The parameters that can be measured from the force-displacement diagram of the fracture experiment of SCB specimen to calculate values of the FI and CRI indices are shown in Figure 7. In this study, the cracking resistance index (CRI) has been measured for the samples at intermediate temperature.

## 5. Laboratory data and results

### 5.1. RV results

Figure 8 represents RV test results at 135 degrees. As can be seen, CLS modified samples have higher viscosity values compared to 60/70 bitumen without additive. In general, with the increase in the CLS percentage, the bitumen viscosity has also increased, and thus, the bitumen containing 20% CLS with a viscosity of 0.75 Pa.s at 135°C has the highest RV among the studied bitumen samples, which shows a 114% increase in the viscosity of bitumen modified with 20% CLS. In the lowest case, when bitumen was mixed with 5% additive, its viscosity reached 0.42, which shows a 20% increase in RV of the modified bitumen. Consequently, CLS material can cause an increment in the stiffness of bitumens in high-temperature condition and increase its viscosity, which is consistent with the research results of Xu [49], Gao [81] and Fatemi [57]. Of course, the important point is that RV results of all bitumens were less than 3 Pa.s. Thus, according to ASTM D4402, which declared the maximum viscosity at 135°C equal to 3 Pa.s, the different percentages of CLS used in this study are acceptable, and the modified bitumens all have appropriate viscosity and workability in the asphalt mixture during pavement implementation, and CLS material inside bitumens does not have an inhibitory effect on the performance of the bitumen.

### 5.2. Low-temperature cracking resistance test results

In this study, two indices of fracture toughness ($K_{1c}$) as well as fracture energy ($G_f$) were used to measure the cracking resistance performance of asphalt mixture containing bitumen modified with CLS at low temperature. The measurement results of $K_{1c}$ index are shown in Figure 9. Based on this, $K_{1c}$ index has also increased for samples with modified bitumen with a maximum of 15% CLS by increasing the additive. In such a way that by increasing the CLS percentage from 0 to 15, the value of $K_{1c}$ has increased by 37%, from 0.38 to 0.521 MPa.$^{m0.5}$. But the samples with 20% modified bitumen not only did not have higher fracture toughness, but compared to the modified sample with 15% CLS, it also decreased by 7.5%. However, compared to the sample with control bitumen, their fracture toughness was 27% higher. As a result, it can be seen that adding CLS to bitumen up to 15% has increased the toughness of the sample and the maximum tolerable load of the samples. But increasing the additive percentage to 20% has reduced the tolerable force of the SCB sample.

As mentioned before, in this article, the fracture energy values of SCB samples of control and modified samples, in addition to the total fracture energy, have been calculated in the form of fracture energy before the critical load (peak point of graph) and after the critical load. The purpose of this work is to determine asphalt mixture behavior against crack formation and propagation. The fracture energy values of different samples are presented in Figure 10. The results of the amount of fracture energy at negative temperature show that the total fracture energy as well as the fracture energy before the critical load for samples containing additive increases gradually with the increase of additive percentage up to 15%. So that the fracture energy of mixtures containing 15% modified bitumen has increased by 55% from 1283 to 1983 J/m$^2$ and the fracture energy before the critical load for the same mixtures has increased by 47% from 1121 to 1646 J/m$^2$. However, with the increase of CLS of bitumen and reaching 20%, the fracture energy not only did not increase, but also decreased, and the total fracture energy decreased by 9% to 1801 J/m$^2$ and the fracture energy before the critical load decreased by 11% to reach 1460 J/m$^2$. The reason for this can be considered to be the increase in bitumen stiffness and consequently the asphalt mixture for 0 to 15 percent CLS and its decrease for 15 to 20 percent CLS. This has also been shown in the studies of Xie [82] and Fatemi [39]. Of course, in general, the fracture toughness and energies of SCB samples of the modified asphalt mixture in this study have always been higher than the control sample. The minimum amount of CLS added to the mixture, which is considered 5% in this research, has caused the total fracture energy and fracture energy until the critical load to increase at negative temperature by 22% and 20%, respectively. Thus, the resistance of mixtures modified by CLS to the formation of cracks at low temperature is higher than the control samples, and its best performance occurred for the mixture containing bitumen modified with 15% CLS, and the increase in the percentage of additive from 15% has a negative effect on its cracking resistance performance.

Moreover, the fracture energy after the critical load of samples at the temperature of -15°C has always been on the rise. Although its growth rate has decreased over time. So that the samples containing 15 and 20% bitumen had almost the same performance in this field. Of course, due to the brittle performance of mixtures at a low temperature and the small area under their force-displacement diagram, the value of this fracture energy is much smaller than the fracture energies that were calculated earlier, and thus, it is not so decisive in determining the behavior of the mixture.

### 5.3. Intermediate temperature cracking resistance test results

In this study, the fracture energy ($G_f$), flexibility index (FI) and cracking resistance index (CRI) parameters were utilized to measure the cracking resistance of mixtures containing bitumen modified with CLS at intermediate temperature. Also, the fracture energy parameter, as mentioned in the low temperature section, is also separated into the fracture energy prior and after critical load.

The fracture energy values of different samples are presented in Figure 11. The fracture energy results at intermediate temperature showed that the total fracture energy as well as before and after the critical load, for the samples containing additive, increase gradually with the increase of additive percentage up to 15%. So that the fracture energy of mixtures containing 15% modified bitumen has increased by 70% from 168 to 286 $J/m^2$ and the fracture energy before critical load for the same mixtures has increased by 80% from 75 to 135 $J/m^2$. But similarly to the negative temperature test, when the bitumen CLS rate exceeds 15% and reaches 20%, the fracture energy not only does not increase, but the total fracture energy decreases by 1% to 283 $J/m^2$ and the fracture energy before the critical load has decreased by 4% to 129 $J/m^2$. This process shows that the presence of CLS in the bitumen of asphalt mixtures increases the stiffness of bitumen and asphalt mixture, which increases its resistance to cracking. Also, this increase in resistance has shown itself in the form of an increase in the amount of maximum load that can be tolerated by the SCB sample and an increase in the amount of ductility and displacement of the place of application of the load until the crack occurs (the peak point of the load). Fatemi et al. also presented a similar result in their research on the rutting resistance of samples containing CLS and found that the addition of up to 15% of CLS to bitumen makes the mixture harder and reduces the depth of rutting, and by increasing the additive percentage from 15%, the process of increasing the stiffness is stopped and the stiffness of the mixture decreases [39]. Although the addition of CLS has generally improved the fracture toughness and energies of the SCB samples of the modified asphalt mixture in all the values investigated in this paper compared to the control sample. Thus, even adding 5% CLS to bitumen has caused the total fracture energy and fracture energy before the critical load to increase at intermediate temperature by 12% and 10%, respectively. Therefore, according to fracture energy results of SCB samples in this research, the resistance of the asphalt mixtures modified with CLS to the formation of cracks at intermediate temperature is higher than that of the control samples, and its best performance occurred for the mixture containing bitumen modified with 15% CLS, and increasing the additive percentage from 15% had a negative effect on its intermediate temperature cracking resistance performance.

Furthermore, the fracture energy after the critical load of samples at a temperature of +15°C has always been on the rise. Although its growth rate has decreased over time. So that the samples containing 15 and 20% bitumen had almost the same performance in this field. Based on the ductility behaviour of asphalts in intermediate temperatures, although with the increase of CLS percentage from 15% to 20%, the amount of maximum load for the sample has decreased, but the amount of ductility of the sample and the displacement of the place of application of the load have increased to such an extent that, in general, the area under the force-displacement curve after the peak point of load, where the resistance of sample versus crack propagations, has increased. Therefore, increasing additive percentages up to 20% has caused the resistance of the sample to the expansion of the cracks created in it to increase with a decreasing trend, and in percentages of 15-20%, the samples had almost the same performance. Of course, this increase in crack propagation resistance can be seen for all the modified samples and even the sample containing 5% additive showed a 15% improvement compared to the control sample.

In addition to the fracture energy index, the FI and CRI indices were also applied for measuring the intermediate temperature cracking behavior, and Figure 12 shows FI index calculation results. It has been observed that the FI index increased from 33.35 for the control sample with an increase in CLS percentage and reached 87.19 in the sample with 15% modified bitumen with a 160% increase. But with the increase of additive percentage, its value has reached 75.83 with a decrease of 13%. As mentioned before, in SCB samples of asphalt mixture, the higher the FI index, it means that the sample shows more resistance to crack propagation. Therefore, it is clear that the asphalt samples modified with bitumen containing CLS have a better performance than the control samples against crack propagation, and this performance improvement has reached its maximum value in the asphalt produced with 15% modified bitumen.

Also, according to the results of the calculation of the CRI index shown in Figure 13, this index has performed completely similar to FI, and the C15 sample has improved the most with an increase of 45% of the index from 1556 to 2258, and after that the index has decreased to some extent. Therefore, by comparing the CRI index of the modified samples with the control, it can be found that the cracking behavior of samples increases by adding CLS, the optimal value of which is around 15 percent by weight of bitumen.

In the intermediate temperature, it can be said that the trend of rising and falling of the graphs for $G_f$, FI and CRI indices was almost the same. But by looking carefully at the values of the indices, we notice that the amount of changes in the FI index is much higher than the other two indices. The reason for this is that the calculation of the FI index is measured with the use of two parameters $G_f$ and m (the slope of the graph in the load return phase), which increases the maximum load by adding CLS to bitumen and increasing the stiffness of the mixture. On the other hand, with the increase in the resistance of the sample against the propagation of cracks, the displacement of the place of load during loading also increases, and this increase causes a decrease in the slope of the graph in the load return phase. Therefore, in the calculation of FI, the numerator of the fraction has increased and its denominator has also decreased, which leads to drastic changes in the calculation of the index.

## 6. Conclusion

The aim of the present research was to evaluate the cracking resistance of asphalt samples containing bitumen modified by calcium lignosulfonate at low and intermediate temperatures. For this purpose, firstly, in order to measure the pumpability and feasibility of workability of 60-70 bitumen without additive, along with modified bitumen with 5, 10, 15 and 20% calcium lignosulfonate by weight of bitumen, rotational viscosity (RV) test was performed on different bitumens. Then, from SCB warm mix asphalt samples made with control and modified bitumens, to measure fracture energy parameters at low and intermediate temperatures (generally and separately for before and after critical load), fracture toughness ($K_{1c}$) was used for negative temperature samples and flexibility index (FI) as well as cracking resistance index (CRI) were used for intermediate temperature samples. Finally, the values of the mentioned indices for mixtures with different bitumens were compared with each other in order to determine the appropriate amount of additive for the best performance of the asphalt mixture modified with calcium lignosulfonate, taking into account its workability. The following results were obtained from this study:

- CLS particles can increase bitumen stiffness in high temperature conditions and increase its viscosity. Thus, the bitumen containing 20% CLS at a temperature of 135°C had a viscosity equal to 0.75 Pa.s.
- RV results of all bitumens at 135°C temperature were less than 3 Pa.s. Therefore, according to ASTM D4402, which declared the maximum viscosity at 135°C equal to 3 Pa.s, the different percentages of CLS used in this study are acceptable, and the modified bitumens all have appropriate viscosity and workability in the asphalt mixture during pavement implementation, and CLS material inside the bitumen does not have an inhibitory effect on the performance of the bitumen.
- As the percentage of CLS increases from 0 to 15%, the s ($K_{1c}$) is increased. But the samples with 20% modified bitumen not only did not have higher fracture toughness, but also had a decrease in CLS compared to the sample modified with 15%. However, compared to the sample with control bitumen, their fracture toughness has been higher. As a result, it can be seen that adding CLS to bitumen up to 15% has increased the stiffness of the sample and the maximum tolerable load of the samples. But increasing the additive percentage to 20% has reduced the tolerable force of the SCB sample.
- Examining the total fracture energy index and pre-critical load in SCB samples showed that the resistance of samples modified with CLS to cracking at low temperatures was higher than that of control samples, and the best performance occurred for the mixture containing bitumen modified with 15% CLS. Increasing the percentage of additive from 15% negatively affects its cracking resistance performance.
- The results of fracture energy before the critical load at intermediate temperature illustrated that the resistance of samples modified with CLS to cracking at intermediate temperature was higher than that of control samples, and its best performance occurred for the mixture containing bitumen modified with 15% CLS, and increasing the additive percentage from 15% negatively affects its intermediate temperature cracking resistance.
- The fracture energy results after the critical load at intermediate temperature showed that increasing the additive percentage up to 20% has caused the resistance of the sample to the expansion of the cracks created in it to increase with a decreasing trend, and at 15-20%, the samples have almost the same performance.

- The FI index values of SCB samples of asphalt mixtures modified with bitumen containing CLS were higher than the control samples, and it shows that the modified asphalt mixture samples have a better performance than the control sample against crack propagation. The FI index increased with the increase of CLS percentage in bitumen up to 15% and then decreased.
- The CRI index also performed quite similar to the FI and its value increased with the addition of CLS to bitumen. But this increase stopped after the CLS percentage exceeded 15% and the index has also decreased. Therefore, the cracking resistance of samples has been increased with adding CLS, the optimal value of which is about 15 percent by weight of bitumen.
- In the intermediate temperature, it can be said that the trend of rising and falling of the graphs for $G_f$, FI and CRI indices was almost the same. But the amount of changes in the FI index is much higher than the other two indices, which is due to the simultaneous increase in the stiffness of the mixture and its fracture energy and the decrease in the slope of the graph in the return phase in samples containing modified bitumen up to 15%.
- Results show that the asphalt mixture modified by CLS additive has its best performance in 15% dosage of additive in low and intermediate temperature
- As mentioned before, FI, CRI, Fracture Energy and Fracture Toughness Index increase by adding CLS up to 15%. But decreasing the CLS percentage to 20% negatively affects its cracking resistance performance. Previous studies declared that more than 15% CLS in asphalt mixture makes it brittle and increases the speed of crack propogation. So 15% is the optimum percentage of CLS additive in asphalt mixture.
- Comparing the result to other studies, Fatemi et al. found that all percentages of CLS have anti-aging properties and also 15% value provides the greatest resistance to creasing [55-57] that is equal with the presented research. also, Zarei et al. investigated various properties of CLS, including cracking resistance, moisture sensitivity in different thaw and freeze cycles, along with the CLS additive and results of these researches showed that CLS can improve the fracture toughness in different temperatures and increases the cracking resistance of the mixture and also improve stripping by increasing toughness [58-61]. The research conducted in this article confirms the results of previous studies on the use of this material as an additive in asphalt mixtures.

**Acknowledgements**

The research of the corresponding author is supported by a grant from Ferdowsi University of Mashhad

**Data Availability**

Based on the logical request of the responsible author, all data supporting the conclusion of this research will be available.

**Conflicts of Interest**

The authors declare that there are no conflicts of interest regarding the publication of this paper.

**Credit authorship contribution statement**

Seyed Ali Ziaee and Seyed Saeed Fatemi: Conceptualization, Methodology, Resources, Supervision, Investigation, and Writing – review & editing.

Seyed Amir Saadatjoo: Investigation, Validation, Visualization, and Writing – review & editing.

Jafar Bolouri Bazaz and Rouzbeh Shad: Response to comments of reviewers, and Writing – review & editing of revised sections.

**References**

1. Dan, H. C., Zhang, Z., Chen, J. et al. "Low-temperature fracture characteristics of asphalt mixtures using the eccentric single-edge notched bend test: A 3D discrete element study", *Construction and Building Materials*, 344, 128182 (2022). https://doi.org/10.1016/j.conbuildmat.2022.128182

2. Shahryari, N., Keymanesh, M. and Aliha, M. "Specimen type effect on measured low-temperature fracture toughness of asphalt concrete", *Fatigue & Fracture of Engineering Materials & Structures*, 44(2): p. 551-567 (2021). https://doi.org/10.1111/ffe.13379

3. Shafabakhsh, G., Sadeghnejad, M. and Ebrahimnia, R. "Fracture resistance of asphalt mixtures under mixed-mode I/II loading at low-temperature: Without and with nano SiO2", *Construction and Building Materials*, 266: p. 120954 (2021). https://doi.org/10.1016/j.conbuildmat.2020.120954

4. Fakhri, M., Kharrazi, E.H. and Aliha, M. "Mixed mode tensile–In plane shear fracture energy determination for hot mix asphalt mixtures under intermediate temperature conditions", *Engineering Fracture Mechanics*, 192: p. 98-113 (2018). https://doi.org/10.1016/j.engfracmech.2018.02.007

5. Zarei, M., Abdi Kordani, A. and Zahedi, M. "Pure mode I and pure mode II fracture resistance of modified hot mix asphalt at low and intermediate temperatures", *Fatigue & Fracture of Engineering Materials & Structures*, 44(8): p. 2222-2243 (2021). https://doi.org/10.1111/ffe.13508

6. Zarei, M., Abdi Kordani, A. and Zahedi, M. "Evaluation of fracture behavior of modified Warm Mix Asphalt (WMA) under modes I and II at low and intermediate temperatures", *Theoretical and Applied Fracture Mechanics*, 114: p. 103015 (2021). https://doi.org/10.1016/j.tafmec.2021.103015

7. Aliha, M., Behbahani, H., Fazaeli, H. et al. "Study of characteristic specification on mixed mode fracture toughness of asphalt mixtures", *Construction and Building Materials*, 54, 623-635 (2014). https://doi.org/10.1016/j.conbuildmat.2013.12.097

8. Karimi, M., Dehaghi, E.A. and Behnood, A. "A fracture-based approach to characterize long-term performance of asphalt mixes under moisture and freeze-thaw conditions", *Engineering Fracture Mechanics*, 241: p. 107418 , (2021). https://doi.org/10.1016/j.engfracmech.2020.107418

9. Aliha, M. and Sarbijan, M. "Effects of loading, geometry and material properties on fracture parameters of a pavement containing top-down and bottom-up cracks", *Engineering Fracture Mechanics*, 166: p. 182-197 (2016).

https://doi.org/10.1016/j.engfracmech.2016.08.028

10. Cannone Falchetto, A., Wistuba, M.P. and Marasteanu, M.O. "Size effect in asphalt mixture at low temperature: Types I and II", *Road Materials and Pavement Design*, 18(sup1): p. 235-257 (2017).

https://doi.org/10.1080/14680629.2016.1266764

11. Ramesh, A., Ramayya, V.V., Reddy, G.S. et al. "Investigations on fracture response of warm mix asphalt mixtures with Nano glass fibres and partially replaced RAP material", *Construction and Building Materials*, 317: p. 126121 (2022).

https://doi.org/10.1016/j.conbuildmat.2021.126121

12. Haghighatpour, P.J. and Aliha, M. "Assessment of freezing and thawing cycle (FTC) effects on mixed mode I/III fracture toughness and work of fracture of HMA asphalt mixtures", *Theoretical and Applied Fracture Mechanics*, 118: p. 103261 (2022).

https://doi.org/10.1016/j.tafmec.2022.103261

13. Bui, H.H. and Saleh, M. "Effects of specimen size and loading conditions on the fracture behaviour of asphalt concretes in the SCB test", *Engineering Fracture Mechanics*, 242: p. 107452 (2021).

https://doi.org/10.1016/j.engfracmech.2020.107452

14. Ameri, M., Mansourian, A., Khavas, M. et al. "Cracked asphalt pavement under traffic loading–A 3D finite element analysis", *Engineering Fracture Mechanics*, 78(8): p. 1817-1826 (2011).

https://doi.org/10.1016/j.engfracmech.2010.12.013

15. Pirmohammad, S. and Majd-Shokorlou, Y. "Finite element analysis of road structure containing top-down crack within asphalt concrete layer", *Journal of Central South University*, 27(1): p. 242-255 (2020).

https://doi.org/10.1007/s11771-020-4292-3

16. Emery, J. and Eng, P. "Mitigation of asphalt pavement top-down cracking", *4th International SIIV Congress*, Palermo, Italy: Società Italiana Infrastrutture Viarie. (2007).

https://www.siiv.net/site/sites/default/files/Documenti/palermo/63_2848_20080108102227.pdf

17. Kim, Y.R., "Modeling of Asphalt Concrete", 1st Edn., New York: ASCE Press ; McGraw-Hill (2009).

https://www.accessengineeringlibrary.com/content/book/9780071464628

18. Du, G. "Implementation of the SuperPave Indirect Tensile Test (IDT) Analysis Procedure", *KTH Architecture and the Built Environment*, 2010: Stockholm, Sweden (2010).

https://vbn.aau.dk/en/publications/implementation-of-the-superpave-indirect-tensile-test-idt-analysi

19. Nsengiyumva, G., You, T., Kim, Y.R. et al. "Investigation of testing variables of semicircular bending test for asphalt concrete mixtures: experimental-statistical approach", *Transportation Research Board 95th Annual Meeting*, Washington DC, United States (2016).

https://trid.trb.org/View/1393706

20. Hoare, T.R. and Hesp, S.A. "Low-temperature fracture testing of asphalt binders: regular and modified systems. *Transportation Research Record*, 1728(1): p. 36-42 (2000).

https://doi.org/10.3141/1728-06

21. Marasteanu, M.O., Dai, S., Labuz, J.F. et al. "Determining the low-temperature fracture toughness of asphalt mixtures", *Transportation Research Record*, 1789(1): p. 191-199 (2002).

https://doi.org/10.3141/1789-21

22. Wagoner, M.P., Buttlar, W.G. and Paulino, G.H. "Development of a single-edge notched beam test for asphalt concrete mixtures", *Journal of Testing and Evaluation*, 33(6): p. 452 (2005).

https://doi.org/10.1520/JTE12579

23. Wagoner, M.P., Buttlar, W.G. and Paulino, G.H. "Disk-shaped compact tension test for asphalt concrete fracture", *Experimental mechanics*, 45(3): p. 270-277 (2005).

https://doi.org/10.1007/BF02427951

24. Wagoner, M.P., Buttlar, W.G., Paulino, G.H. et al. et al. “Laboratory testing suite for characterization of asphalt concrete mixtures obtained from field cores. in Asphalt Paving Technology”, *Association of Asphalt Paving Technologists-Proceedings of the Technical Sessions*. Association of Asphalt Paving Technologist (2006).

http://worldcat.org/issn/02702932

25. Aliha, M., Bahmani, A. and Akhondi, S. “Numerical analysis of a new mixed mode I/III fracture test specimen”, *Engineering Fracture Mechanics*, 134: p. 95-110 (2015).

https://doi.org/10.1016/j.engfracmech.2014.12.010

26. Lim, I., Johnston, I. and Choi, S. “Stress intensity factors for semi-circular specimens under three-point bending”, *Engineering Fracture Mechanics*, 44(3): p. 363-382 (1993).

https://doi.org/10.1016/0013-7944(93)90030-V

27. Aliha, M., Fazaeli, H., Aghajani, S. et al. “Effect of temperature and air void on mixed mode fracture toughness of modified asphalt mixtures”, *Construction and Building Materials*, 95: p. 545-555 (2015).

https://doi.org/10.1016/j.conbuildmat.2015.07.165

28. Fakhri, M., Haghighat Kharrazi, E., Aliha, M. et al. “The effect of loading rate on fracture energy of asphalt mixture at intermediate temperatures and under different loading modes”, *Frattura ed Integrità Strutturale*, 12(43): p. 113-132 (2018).

https://doi.org/10.3221/IGF-ESIS.43.09

29. Ameri, M., Mansourian, A., Pirmohammad, S. et al. “Mixed mode fracture resistance of asphalt concrete mixtures”, *Engineering Fracture Mechanics*, 93: p. 153-167 (2012).

https://doi.org/10.1016/j.engfracmech.2012.06.015

30. Chong, K.P. and Kuruppu, M.D. “New specimen for fracture toughness determination for rock and other materials”, *International Journal of Fracture*, 26(2): p. R59-R62 (1984).

https://doi.org/10.1007/BF01157555

31. Pirmohammad, S. and Kiani, A. “Study on fracture behavior of HMA mixtures under mixed mode I/III loading”, *Engineering Fracture Mechanics*. 153: p. 80-90 (2016).

https://doi.org/10.1016/j.engfracmech.2015.12.027

32. Pirmohammad, S., Majd-Shokorlou, Y. and Amani, B. “Experimental investigation of fracture properties of asphalt mixtures modified with Nano Fe2O3 and carbon nanotubes”, *Road Materials and Pavement Design*, 21(8): p. 2321-2343 (2020).

https://doi.org/10.1080/14680629.2019.1608289

33. Zofka, A. and Braham, A. “Comparison of low-temperature field performance and laboratory testing of 10 test sections in the Midwestern United States”, *Transportation research record*, 2127(1): p. 107-114 (2009).

https://doi.org/10.3141/2127-13

34. Al-Qadi, I. L., Ozer, H., Lambros, J. et al. “Testing protocols to ensure performance of high asphalt binder replacement mixes using RAP and RAS”, *Illinois Center for Transportation/Illinois Department of Transportation*. No. 15-017 (2015).

https://hdl.handle.net/2142/88680

35. Im, S., Ban, H. and Kim, Y. “Characterization of mode-I and mode-II fracture properties of fine aggregate matrix using a semicircular specimen geometry”, *Construction and Building Materials*, 52: p. 413-421 (2014).

https://doi.org/10.1016/j.conbuildmat.2013.11.055

36. Wu, Z., Mohammad, L.N., Wang, L.B. et al. “Fracture resistance characterization of superpave mixtures using the semi-circular bending test”. *ASTM International* (2005).

https://doi.org/10.1520/JAI12264

37. Ameri, M., Mohammadi, R., Mousavinezhad, M. et al. “Evaluating Properties of Asphalt Mixtures Containing polymers of Styrene Butadiene Rubber (SBR) and recycled Polyethylene Terephthalate (rPET) against Failures Caused by Rutting, Moisture and Fatigue”. *Frattura ed Integrità Strutturale*, 14(53): p. 177-186 (2020).

https://doi.org/10.3221/IGF-ESIS.53.15

38. Wang, R., Xiong, Y., Yue, M. et al. "Investigating the effectiveness of carbon nanomaterials on asphalt binders from hot storage stability, thermodynamics, and mechanism perspectives", *Journal of Cleaner Production*, 276: p. 124180 (2020).
https://doi.org/10.1016/j.jclepro.2020.124180

39. Fatemi, S., Bazaz, J.B and Ziaee, S.A. "Investigating the effect of calcium lignosulfonate on the durability and performance of asphalt mixtures. *Advances in Materials Science and Engineering*, 5260159 (2022).
https://doi.org/10.1155/2022/5260159

40. Qian, G., Yang, C., Huang, H. et al. "Resistance to ultraviolet aging of nano-SiO2 and rubber powder compound modified asphalt", *Materials*, 13(22): p. 5067 (2020).
https://doi.org/10.3390/ma13225067

41. Liu, W., Xu, Y., Wang, H. et al. "Enhanced storage stability and rheological properties of asphalt modified by activated waste rubber powder", *Materials*, 14(10): p. 2693 (2021).
https://doi.org/10.3390/ma14102693

42. Li, H., Li, W., Sheng, Y. et al. "Influence of compound action of rubber powder and SBS on high-temperature performance of asphalt pavement surface", *Journal of Materials in Civil Engineering*, 33(6): p. 04021126 (2021).
https://doi.org/10.1061/(ASCE)MT.1943-5533.0003718

43. Zarei, M., Rahmani, Z., Zahedi, M. et al. "Technical, economic, and environmental investigation of the effects of rubber powder additive on asphalt mixtures", *Journal of Transportation Engineering, Part B: Pavements*, 146(1): p. 04019039 (2020).
https://doi.org/10.1061/JPEODX.0000142

44. Arafat, S., Kumar, N., Wasiuddin, N.M. et al. "Sustainable lignin to enhance asphalt binder oxidative aging properties and mix properties", *Journal of Cleaner Production*, 217: p. 456-468 (2019).
https://doi.org/10.1016/j.jclepro.2019.01.238

45. Gilca, I.A., Ghitescu, R.E., Puitel, A.C. et al. “Preparation of lignin nanoparticles by chemical modification”, *Iranian polymer journal*, 23(5): p. 355-363 (2014). https://doi.org/10.1007/s13726-014-0232-0

46. Ouyang, X., Qiu, X. and Chen, P. “Physicochemical characterization of calcium lignosulfonate—a potentially useful water reducer”, *Colloids and Surfaces A: Physicochemical and Engineering Aspects*, 282: p. 489-497 (2006). https://doi.org/10.1016/j.colsurfa.2005.12.020

47. Behin, J. and Sadeghi, N. “Utilization of waste lignin to prepare controlled-slow release urea”, *International Journal of Recycling of Organic Waste in Agriculture*, 5(4): p. 289-299 (2016). https://doi.org/10.1007/s40093-016-0139-1

48. Zhu, F., Li, J., Jiang, W. et al. “Freeze-thaw performance of silt sand treated with lignin”, *Advances in Civil Engineering*, 2021: p. 1-10 (2021). https://doi.org/10.1155/2021/6639268

49. Xu, G., Wang, H. and Zhu, H. “Rheological properties and anti-aging performance of asphalt binder modified with wood lignin”, *Construction and Building Materials*, 151: p. 801-808 (2017). https://doi.org/10.1016/j.conbuildmat.2017.06.151

50. Tribot, A., Amer, G., Alio, M.A. et al. “Wood-lignin: Supply, extraction processes and use as bio-based material”, *European Polymer Journal*, 112: p. 228-240 (2019). https://doi.org/10.1016/j.eurpolymj.2019.01.007

51. Qiu, X., Yan, M., Yang, D. et al. “Effect of straight-chain alcohols on the physicochemical properties of calcium lignosulfonate”, *Journal of colloid and interface science*, 338(1): p. 151-155 (2009). https://doi.org/10.1016/j.jcis.2009.05.072

52. Yu, J., Vaidya, M., Su, G. et al. “Experimental study of soda lignin powder as an asphalt modifier for a sustainable pavement material”, *Construction and Building Materials*, 298: p. 123884 (2021).

https://doi.org/10.1016/j.conbuildmat.2021.123884

53. Batista, K.B., Padilha, R., Castro, T.O. et al. “High-temperature, low-temperature and weathering aging performance of lignin modified asphalt binders”, *Industrial Crops and Products*, 111: p. 107-116 (2018).

https://doi.org/10.1016/j.indcrop.2017.10.010

54. McCready, N.S. and Williams, R.C. “Utilization of biofuel coproducts as performance enhancers in asphalt binder”, *Transportation research record*, 2051(1): p. 8-14 (2008).

https://doi.org/10.3141/2051-02

55. Fatemi, S., Bolouri, J. and Ziaee, S.A. “The pros and cons of using calcium lignosulfonate as a recycled anti-aging additive on engineering properties of bituminous mastics”, *Case Studies in Construction Materials*, 15: p. e00739 (2021).

https://doi.org/10.1016/j.cscm.2021.e00739

56. Fatemi, S., Bolouri, J. and Ziaee, S.A. “Evaluation of rutting and fatigue behaviors of asphalt binders modified with calcium lignosulfonate”, *Advances in Civil Engineering*, 2021: p. 1-12 (2021).

https://doi.org/10.1155/2021/6894514

57. Fatemi, S., Bolouri, J. and Ziaee, S.A. “Laboratory Investigation of Using Calcium Lignosulfonate as an Oxidation Inhibitor in Bitumen”, *Advances in Civil Engineering*, 1488958 (2022).

https://doi.org/10.1155/2022/1488958

58. Zarei, M., Kordani, A.A., Zahedi, M. et al. “Evaluation of low and intermediate temperatures fracture indices for modified Warm mix asphalt (WMA) using edge notched disc bend (ENDB) specimen”, *Theoretical and Applied Fracture Mechanics*, 116: p. 103137 (2021).

https://doi.org/10.1016/j.tafmec.2021.103137

59. Zarei, M., Kordani, A.A., Khanjari, M. et al. “Evaluation of fracture resistance of asphalt concrete involving Calcium Lignosulfonate and Polyester fiber under freeze–thaw damage”, *Theoretical and Applied Fracture Mechanics*, 117: p. 103168 (2022). https://doi.org/10.1016/j.tafmec.2021.103168

60. Zarei, M., Kordani, A.A., Naseri, A. et al. “Evaluation of fracture behaviour of modified warm mix asphalt containing vertical and angular cracks under freeze-thaw damage”, *International Journal of Pavement Engineering*, p. 1-17 (2022). https://doi.org/10.1080/10298436.2022.2072500

61. Zarei, M., Kordani, A.A. and Zahedi, M. “Evaluating the fracture behaviour of modified asphalt concrete composites (ACC) at low and intermediate temperatures using edge notched disc bend (ENDB) specimen”, *Road Materials and Pavement Design*, 23(8): p. 1917-1941 (2022). https://doi.org/10.1080/14680629.2021.1950819

62. Fatemi, S. and Imaninasab, R. “Performance evaluation of recycled asphalt mixtures by construction and demolition waste materials”, *Construction and building materials*, 120: p. 450-456 (2016). https://doi.org/10.1016/j.conbuildmat.2016.05.117

63. Aflaki, S. and Tabatabaee, N. “Proposals for modification of Iranian bitumen to meet the climatic requirements of Iran”, *Construction and Building Materials*, 23(6): p. 2141-2150 (2009). https://doi.org/10.1016/j.conbuildmat.2008.12.014

64. Zhang, Y., Liu, X., Apostolidis, P. et al. “Chemical and rheological evaluation of aged lignin-modified bitumen”, *Materials*, 12(24): p. 4176 (2019). https://doi.org/10.3390/ma12244176

65. Jędrzejczak, P., Collins, M.N., Jesionowski, T. et al. "The role of lignin and lignin-based materials in sustainable construction–a comprehensive review", *International Journal of Biological Macromolecules*, 187: p. 624-650 (2021).
https://doi.org/10.1016/j.ijbiomac.2021.07.125

66. Lin, Y., Hu, C., Adhikari, S. et al. "Evaluation of waste express bag as a novel bitumen modifier", *Applied Sciences*, 9(6): p. 1242 (2019).
https://doi.org/10.3390/app9061242

67. Ahmedzade, P., Fainleib, A., Günay, T. et al. "Modification of bitumen by electron beam irradiated recycled low density polyethylene", *Construction and Building Materials*, 69: p. 1-9 (2014).
https://doi.org/10.1016/j.conbuildmat.2014.07.027

68. Mirsepahi, M., Tanzadeh, J. and Ghanoon, S.A. "Laboratory evaluation of dynamic performance and viscosity improvement in modified bitumen by combining nanomaterials and polymer", *Construction and Building Materials*, 233: p. 117183 (2020).
https://doi.org/10.1016/j.conbuildmat.2019.117183

69. Ameri, M., Vamegh, M., Imaninasab, R. et al. "Effect of nanoclay on performance of neat and SBS-modified bitumen and HMA", *Petroleum Science and Technology*,. 34(11-12): p. 1091-1097 (2016).
https://doi.org/10.1080/10916466.2016.1163394

70. Aliha, M., Bahmani, A. and Akhondi, S. "A novel test specimen for investigating the mixed mode I+ III fracture toughness of hot mix asphalt composites–Experimental and theoretical study", *International Journal of Solids and Structures*, 90: p. 167-177 (2016).
https://doi.org/10.1016/j.ijsolstr.2016.03.018

71. Amirdehi, H. F., Aliha, M. Moniri, A. et al. "Using the generalized maximum tangential stress criterion to predict mode II fracture of hot mix asphalt in terms of mode I results–A statistical analysis", *Construction and Building Materials*,. 213: p. 483-491 (2019).
https://doi.org/10.1016/j.conbuildmat.2019.04.067

72. Ayatollahi, M. and Aliha, M. “Wide range data for crack tip parameters in two disc-type specimens under mixed mode loading”, *Computational materials science*, 38(4): p. 660-670 (2007).

https://doi.org/10.1016/j.commatsci.2006.04.008

73. Saha, G. and Biligiri, K.P. “Fracture damage evaluation of asphalt mixtures using Semi-Circular Bending test based on fracture energy approach”, *Engineering Fracture Mechanics*, 142: p. 154-169 (2015).

https://doi.org/10.1016/j.commatsci.2006.04.008

74. Minhajuddin, M., Saha, G. and Biligiri, K.P. “Crack propagation parametric assessment of modified asphalt mixtures using linear elastic fracture mechanics approach”, *Journal of Testing and Evaluation*, 44(1) (2016).

https://doi.org/10.1520/JTE20140510

75. Kuruppu, M. D., Obara, Y., Ayatollahi, M et al. “ISRM-suggested method for determining the mode I static fracture toughness using semi-circular bend specimen”, *Rock Mechanics and Rock Engineering*, 47: p. 267-274 (2014).

https://doi.org/10.1007/s00603-013-0422-7

76. Ziari, H. and Moniri, A. “Laboratory evaluation of the effect of synthetic Polyolefin-glass fibers on performance properties of hot mix asphalt”, *Construction and Building Materials*, 213: p. 459-468 (2019).

https://doi.org/10.1016/j.conbuildmat.2019.04.084

77. Biligiri, K.P., Said, S. and Hakim, H. “Asphalt Mixtures' Crack Propagation Assessment using Semi-Circular Bending Tests”, *International Journal of Pavement Research & Technology*, 5(4) (2012).

http://www.ijprt.org.tw/reader/pdf.php?id=239

78. Ozer, H., Al-Qadi, I.L., Singhvi, P. et al. “Fracture characterization of asphalt mixtures with high recycled content using Illinois semicircular bending test method and flexibility index”, *Transportation Research Record*, 2575(1): p. 130-137 (2016).

https://doi.org/10.3141/2575-14

79. Zhou, F., Im, S., Sun, L. et al. "Development of an IDEAL cracking test for asphalt mix design and QC/QA", *Road Materials and Pavement Design*, 18(sup4): p. 405-427 (2017). https://doi.org/10.1080/14680629.2017.1389082

80. Kaseer, F., Yin, F., Arámbula-Mercado, E. et al. "Development of an index to evaluate the cracking potential of asphalt mixtures using the semi-circular bending test", *Construction and Building Materials*, 167: p. 286-298 (2018). https://doi.org/10.1016/j.conbuildmat.2018.02.014

81. Gao, J., Wang, H., Liu, C. et al. "High-temperature rheological behavior and fatigue performance of lignin modified asphalt binder", *Construction and Building Materials*,. 230: p. 117063 (2020). https://doi.org/10.1016/j.conbuildmat.2019.117063

82. Shangxian, X., Qiang, L., Pravat, K. et al. "Lignin as renewable and superior asphalt binder modifier", *ACS sustainable chemistry & engineering*, 5(4): p. 2817-2823 (2017). https://doi.org/10.1021/acssuschemeng.6b03064

**Tables captions:**



**Figures captions:**

Figure 1. Grading of aggregates

Figure 2. CLS additive

Figure 3. A sample of bitumen in additive mixing

Figure 4. Rotational viscosity measuring device

Figure 5. UTM device for static loading of samples

Figure 6. Force-displacement diagram as well as fracture energy before and after the fracture point (critical load)

Figure 7. Measurable parameters from the force-displacement diagram to calculate the values of FI and CRI indices

Figure 8. RV test results in bitumen samples at 135°C

Figure 9. The results of the calculation of fracture toughness ($K_{1c}$) at -15°C

Figure 10. The results of the calculation of fracture energy ($G_f$) at -15°C, (a) total fracture energy, (b) fracture energy before critical load and (c) fracture energy after critical load

Figure 11. The results of the calculation of fracture energy ($G_f$) at +15°C, (a) total fracture energy, (b) fracture energy before critical load and (c) fracture energy after critical load

Figure 12. The results of the calculation of flexibility index (FI) of SCB samples at +15 temperature

Figure 13. The results of the calculation of cracking resistance index (CRI) of SCB samples at +15 temperature

**Tables:**

Table 1. Physical characteristics of aggregates

| **Test** | **Unit** | **ASTM Standard** | **Test Result** | **Specification Limit** |
|---|---|---|---|---|
| **Coarse Aggregate** | | | | |
| Los Angeles Abrasion Value | % | C131 | 21 | ≤ 25 |
| Fractured Particles in One Side | % | D5821 | 96 | ≥ 95 |
| Fractured Particles in Two Sides | % | D5821 | 92 | ≥ 90 |
| Bulk Specific Gravity | $gr/m^3$ | C127 | 2.66 | - |
| Apparent Specific Gravity | $gr/m^3$ | C127 | 2.74 | - |
| Water Absorption | % | C127 | 0.84 | ≤ 2.5 |
| Fine Aggregate | | | | |
| Bulk specific gravity | $gr/m^3$ | C128 | 2.66 | - |
| Apparent Specific Gravity | $gr/m^3$ | C128 | 2.87 | - |
| Water Absorption | % | C128 | 3.3 | ≤ 2.5 |
| Sand Equivalent | % | D2419 | 74 | ≥ 50 |

Table 2. Physical characteristics of bitumen

| Test Properties | Unit | ASTM Standard | Test Result |
|---|---|---|---|
| Penetration @ 25℃ | 0.1 mm | D5 | 67 |
| Softening Point | ℃ | C36 | 51 |
| Viscosity @ 135℃ | Pa.s | - | 0.354 |
| Specific Gravity | gr/cm$^3$ | D70 | 1.02 |
| Ductility @ 25℃ | cm | D113 | 100+ |
| Flash Point | ℃ | D92 | 280 |
| PG | - | - | 64-16 |

Table 3. Chemical compounds in calcium lignosulfonate

| Constituent | $Na_2O$ | MgO | $K_2O$ | $SO_3$ | CaO | $Fe_2O_3$ | $P_2O_5$ | $TiO_2$ | MnO | LOI(950℃) |
|---|---|---|---|---|---|---|---|---|---|---|
| Wt(%) | 0.04 | 0.17 | 0.12 | 2.9 | 8.0 | 0.08 | 0.11 | 0.01 | 0.08 | 88.29 |

Table 4. Physical characteristics of calcium lignosulfonate

| **Test** | **Color** | **Dry Matter** | **pH (10% solution)** | **Insolubility (v/v)** | **Bulk Density** | **Reducing Sugars** |
|---|---|---|---|---|---|---|
| **Unit** | - | % min | - | % max | $Kg/m^3$ | % |
| **Test Result** | Light Brown | 93.0 | 7.5 ± 0.8 | 0.5 | 500 | 3 |

Table 5. Conditions of the Superpave gyratory compactor for compaction of asphalt mixtures

| **Test** | **Aggregate Temperature (℃)** | **No. of Gyration** | **Stress Level (KPa)** | **Mold Rotation (degree)** | **Angle (degree)** | **Sample Diameter (mm)** | **Air Void (%)** |
|---|---|---|---|---|---|---|---|
| **Specifications** | 160 | 60-200 | 600 | 360 | 1.25 | 150 | 6 |

Table 6. ID and characteristics of asphalt mixtures

| **Sample ID** | **Polymer (%)** | **Thickness (mm)** |
|---|---|---|
| **C0+15** | 0 | |
| **C5+15** | 5 | |
| **C10+15** | 10 | 30 |
| **C15+15** | 15 | |
| **C20+15** | 20 | |
| **C0-15** | 0 | |
| **C5-15** | 5 | |
| **C10-15** | 10 | 27 |
| **C15-15** | 15 | |
| **C20-15** | 20 | |

Table 7. Bitumen samples for RV test

| **Sample ID** | **C-0** | **C-5** | **C-10** | **C-15** | **C-20** |
|---|---|---|---|---|---|
| **Polymer (percentage)** | 0 | 5 | 10 | 15 | 20 |

**Figures:**

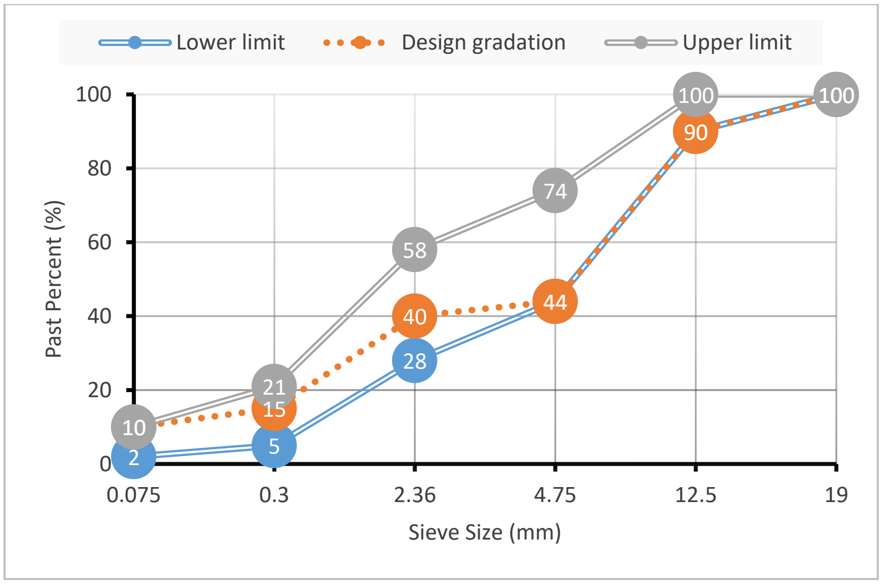



Figure 1. Grading of aggregates

Figure 2. CLS additive

Figure 3. A sample of bitumen in additive mixing

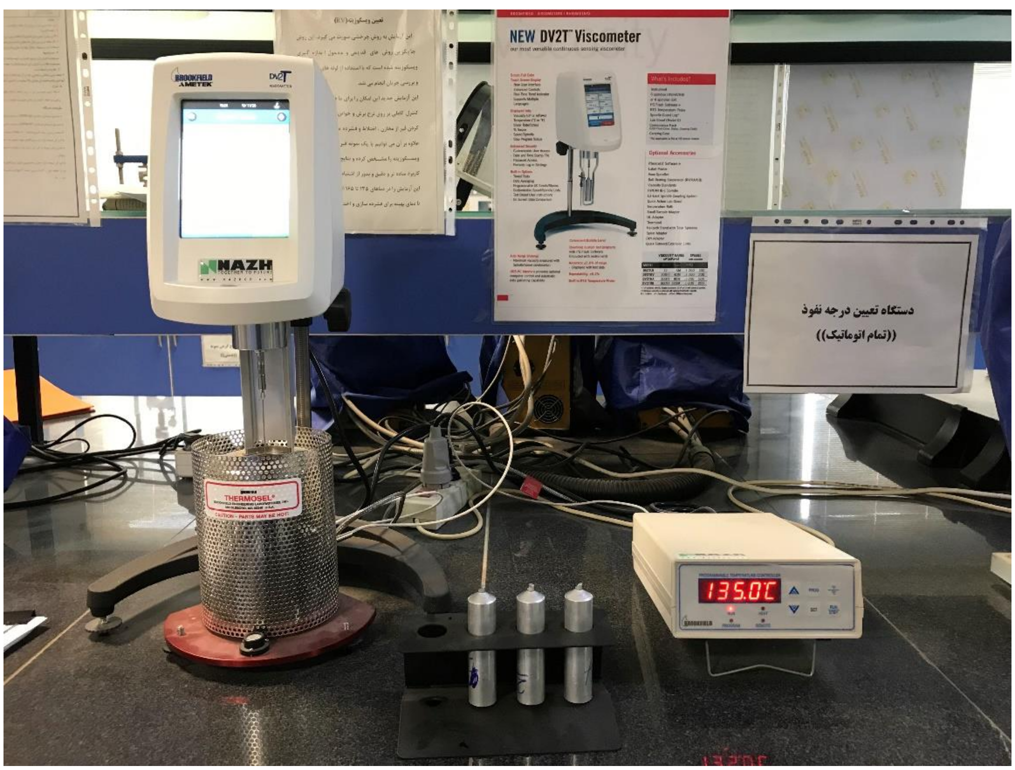



Figure 4. Rotational viscosity measuring device


Figure 5. UTM device for static loading of samples

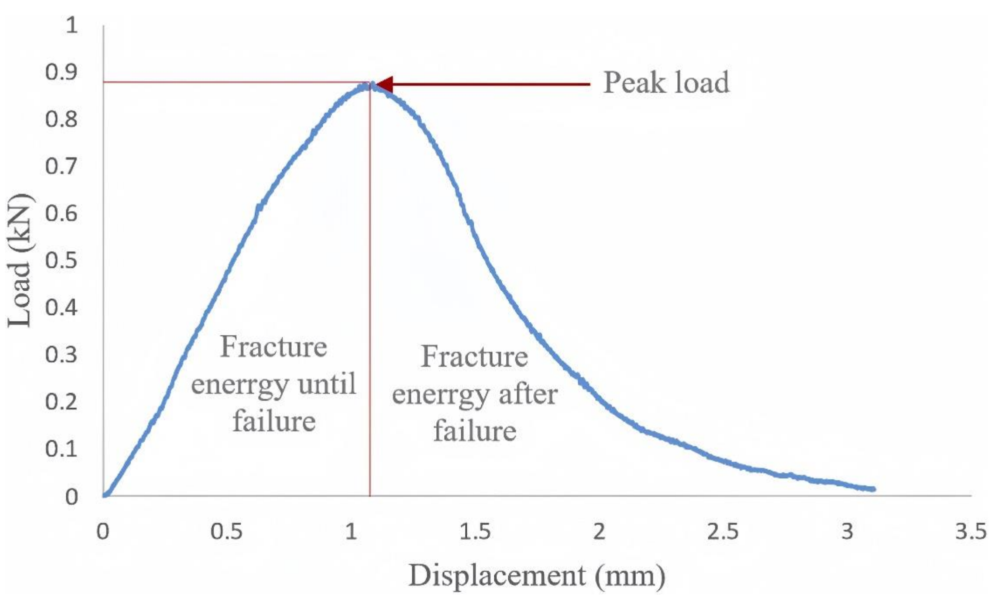



Figure 6. Force-displacement diagram as well as fracture energy before and after the fracture point (critical load)

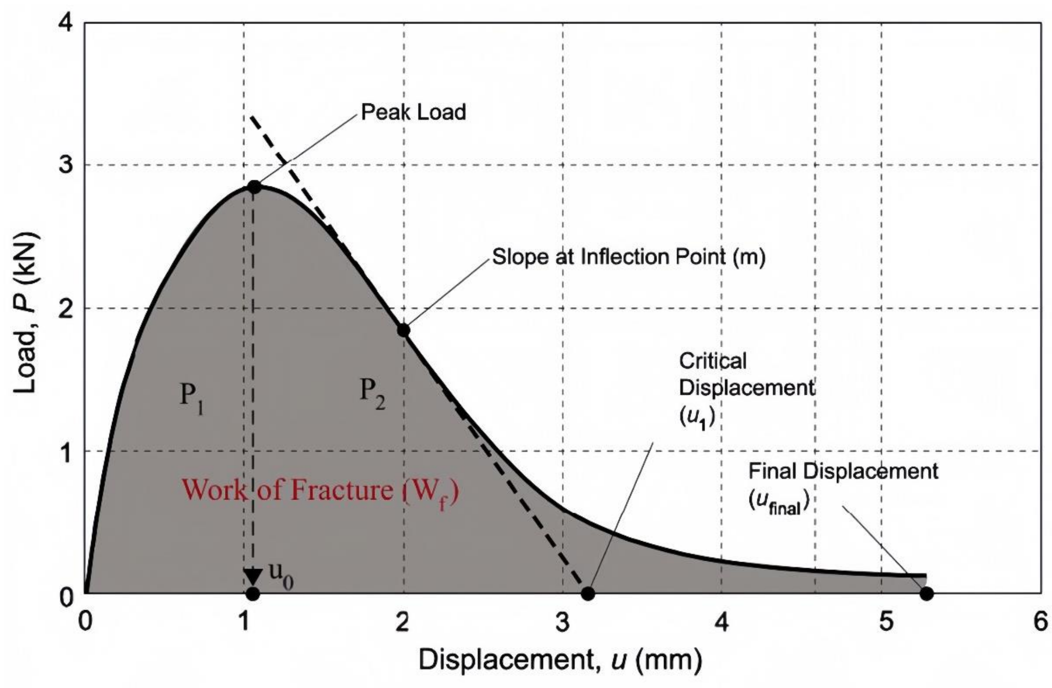



Figure 7. Measurable parameters from the force-displacement diagram to calculate the values of FI and CRI indices

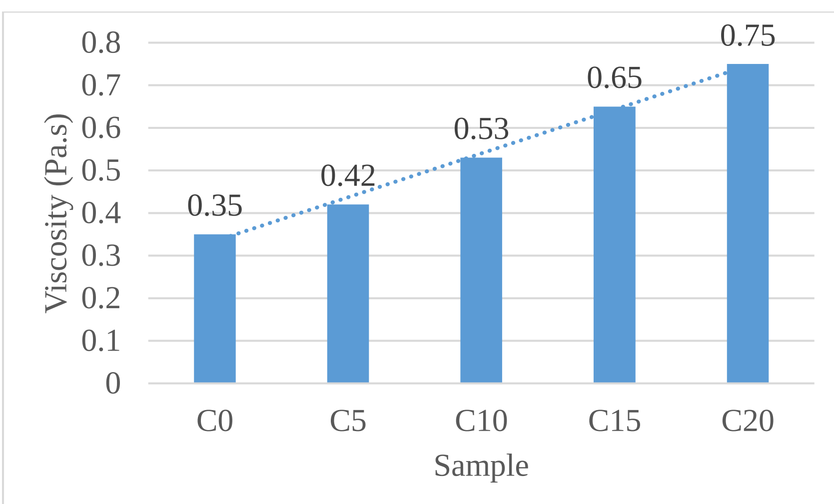



Figure 8. RV test results in bitumen samples at 135°C

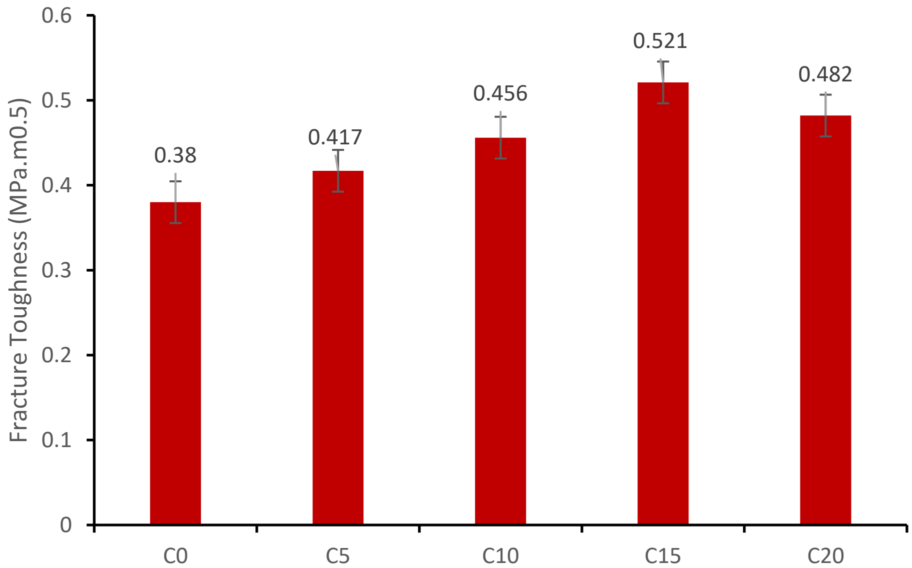
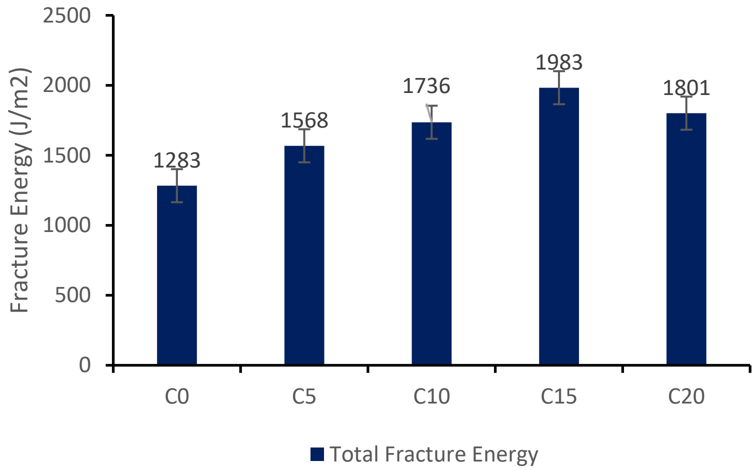
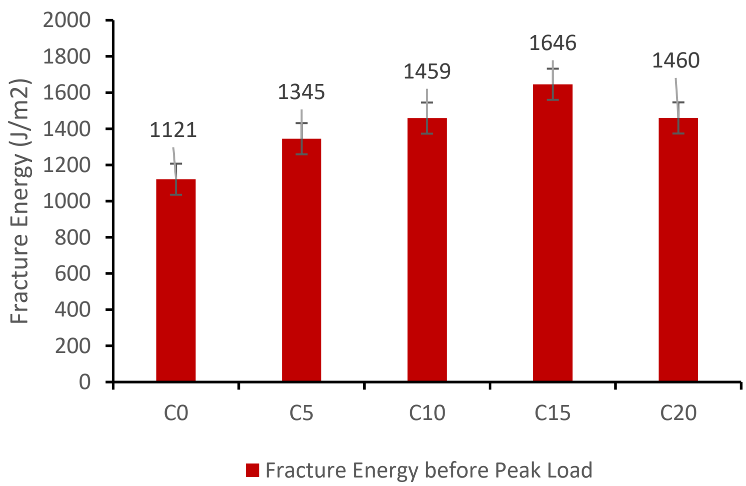



Figure 9. The results of the calculation of fracture toughness ($K_{1c}$) at -15°C

(a)




(b)




(c)

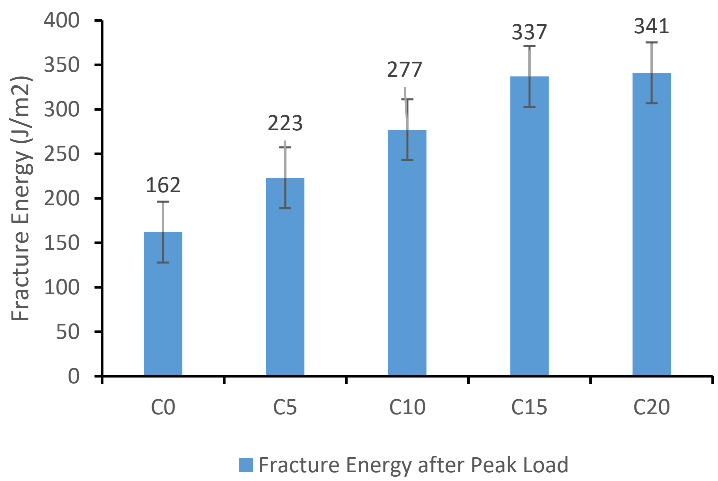
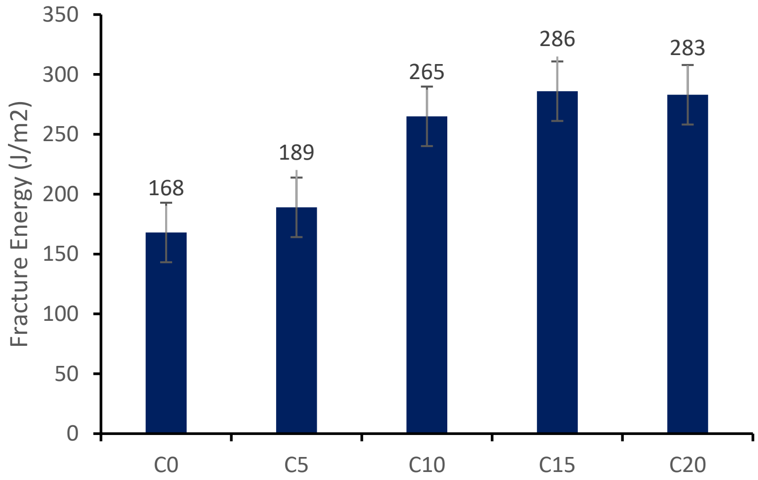
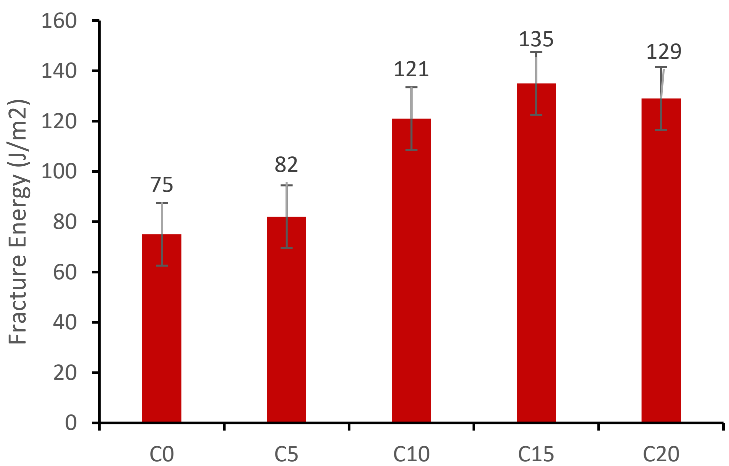



Figure 10. The results of the calculation of fracture energy ($G_f$) at -15°C, (a) total fracture energy, (b) fracture energy before critical load and (c) fracture energy after critical load

(a)




(b)




(c)

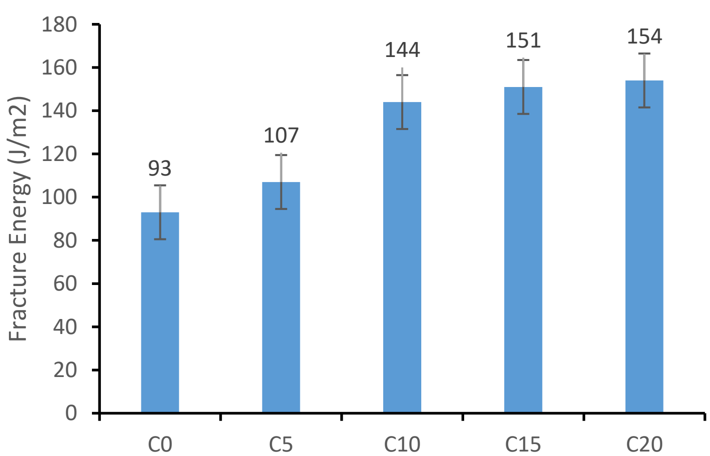



Figure 11. The results of the calculation of fracture energy ($G_f$) at +15°C, (a) total fracture energy, (b) fracture energy before critical load and (c) fracture energy after critical load

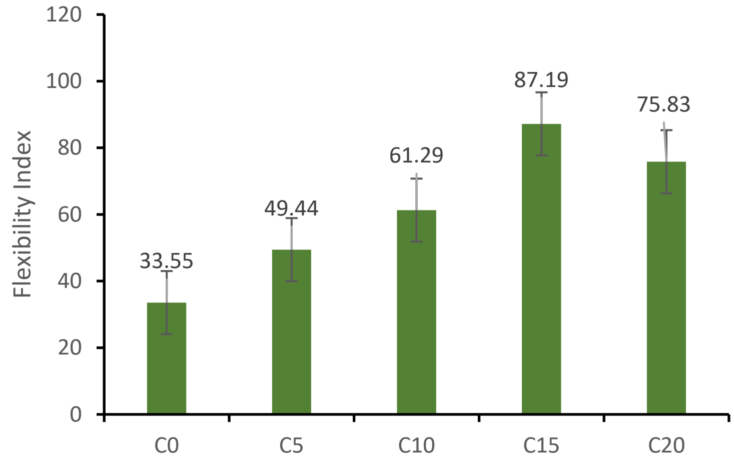



Figure 12. The results of the calculation of flexibility index (FI) of SCB samples at +15 temperature

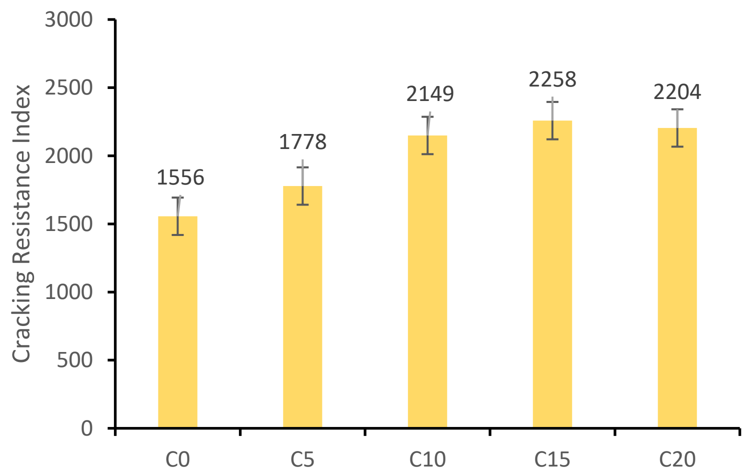



Figure 13. The results of the calculation of cracking resistance index (CRI) of SCB samples at +15 temperature

Seyed Ali Ziaee: I am graduated with a PhD degree from the Sharif University. My focus is on research projects related to asphalt, bitumen, and concrete such as creep, fracture, and water sensitivity. My research outputs have been published in high-level journals. I am a faculty member of Ferdowsi University of Mashhad and I work as an assistant professor at the Ferdowsi University of Mashhad.

Seyed Saeed Fatemi: I am graduated with a PhD degree from the Ferdowsi University of Mashhad. My focus is on research projects related to asphalt, bitumen, and concrete such as creep, fracture, and water sensitivity. My research outputs have been published in high-level journals.

Roozbeh Shad: I am graduated with a PhD degree from the Khajeh Nasir University. My focus is on research projects related to GIS, traffic, and pavements. My research outputs have been published in high-level journals. I am a faculty member of Ferdowsi University of Mashhad and I work as an associate professor at the Ferdowsi University of Mashhad.

Jafar Bolouri Bazzaz: I am graduated with a PhD degree from the King's College London. My focus is on research projects related to soil, stabilization, and pavements. My research outputs have been published in high-level journals. I am a faculty member of Ferdowsi University of Mashhad and I work as an associate professor at the Ferdowsi University of Mashhad.

Seyed Amir Saadatjoo: A am a PhD candidate in Civil Engineering Department at Iran University of Science and Technology in Tehran. My focus is on research projects related to asphalt, bitumen, and concrete such as fracture, fatigue and water sensitivity. My research outputs have been published in high-level journals.