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# THE INFLUENCE OF CHEMICAL COMPOSITION OF ASPHALT CEMENT ON THE PHYSICAL AND RHEOLOGICAL PROPERTIES

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**Abstract**

Failure in asphalt mixture and distress in pavement are major issues to roads infrastructure. Selecting an appropriate chemical composition of asphalt cement is a key component in avoiding these issues. This work aimed to investigate the effect of the chemical composition of different polar fractions on the rheological and physical properties of asphalt cement. Four types of asphalt cement with penetration grades of 20/30, 40/50, 60/70 and 85/100 were divided into four fractions. Complex shear modules, rutting resistance and rotational viscosity of the asphalt cement were determined by using a Dynamic Shear Rheometer and a Rotation Viscometer, respectively. The results show that an increase in the asphaltene content and Gastel index resulted in an increase in the complex shear modulus, rutting resistance and rotation viscosity of the asphalt cement. The addition of more asphaltene content and Gastel index resulted in a decrease in penetration and ductility values. This observation also revealed that asphalt cement with higher asphaltene content had higher stiffness. The findings from this study can assist in the understanding of the behaviour of asphalt cements in its original state and improve the performance of asphalt cement for pavement applications.

Keywords: Asphalt cement, Chemical composition, Physical properties, Rheological properties.

## 1. Introduction

Studying the chemical composition of asphalt cement and its impacts on rheological and physical properties can improve the properties of asphalt from refineries, which has led to make a more economic system and lower impact on the environment. Chemical complexity is a problem inherent in studying asphalt cement composition, which renders impossible, the separation of asphalt cement into chemically pure components. Consequently, chemical studies have been limited to group separations, for which group membership depends on the source of the original unrefined petroleum and the assembling procedure in the processing plant [1].

Asphalt cements can be separated into two major fractions: asphaltenes and maltenes [2]. The physical characteristics of an asphalt cement are highly affected by the asphaltene content. Increasing asphaltene content results in stiffer and more sticky asphalt cement with low penetration and ductility, and high viscosity and softening point [3]. While maltenes consist of saturates, aromatics and resins. Depending on the quantity of the four fractions, Saturates, Aromatics, Resins and Asphaltene (SARA), asphalt cement is divided into gel and a sol type. The sol type displays an adequate quantity of aromatics and resins of a sufficient solvating power, whereas the asphaltenes are completely peptised and the subsequent micelles have free movements inside the asphalt cement. For the gel type, the aromatic and resin fractions are not presented in adequate amounts to peptise the micelles, or have not sufficient solvating power, whereas the asphaltenes associate and form irregular structures. This may lead to an unpredictable open packed structure of connected micelles in which the inner voids are full of a liquid of a mixed constitution. Gel type asphalt cement is less ductile and more elastic than the sol type. Most asphalt cements have intermediate character [1, 4].

In addition to the contents of (SARA) fractions, various indices were considered such as the Gastel index (*Ic*) which is defined as the dispersing capability of maltene to asphaltene and noted as the peptizing power, see Eq. (1) [5]. Oyekunle [6] demonstrated that the growth of the Gastel index leads to a decrease in colloidal stability.

$$Ic = \frac{(Saturate + Asphaltene)}{(Aromatic + Resin)} \tag{1}$$

Another indicator was the ratio of aromatic to asphaltene (*Iar/as*), which permits an assessment of the solvation capacity of the asphalt cement. Decreasing *Iar/as* ratio indicates fewer peptised asphaltenes, and results in increasing the gel character and the softening point of the asphalt cement [1, 7].

In the literature, several studies have been conducted on the influence of chemical composition on rheological and physical properties. For example, Weigel and Stephan [8] studied the relationship between the softening point and the asphaltene content, the found that increasing the asphaltene content results in enhancing the softening point. Due to the straight relationship between the viscosity and the softening point of the asphalt cement, a higher asphaltene content results in increasing the viscosity of the asphalt cement.

Villacorta et al. [9] demonstrated that the rheology of asphalt cement relies upon the combination of the dissimilar constitutive fractions and significantly relies upon the grade of the connotation of asphaltenes and the relative amount of other substances present in the system to stabilize those associations. In other words, the rheology of the asphalt cement is extremely affected by the grade to which the resins are active in saving the asphaltene dispersed in the oil.

Other researchers such as Hermadi et al. [10] studied the effect of saturates on the rheological properties of asphalt cement, and the results showed that the rise of saturates lead to the decrease in all the rheological properties of the asphalt cement and became more resistant to fatigue cracking, but less elastic, viscous, and resistant to plastic failure.

Hermadi and Pravianto [11] described the effect of resins on rheological characteristics of asphalt cement. The results indicated that there was no difference in the effect of the experiments using different types of asphalt cement (rock asphalt or petroleum asphalt) on the complex shear modulus $G^*$ and phase angle $\delta$. However, the percentage of resins, test temperature, and ageing condition have an effect on $G^*$ and $\delta$.

The rheological properties of asphalt cement in this work related to the superpave system. The superpave system is planned to progress the behaviour of asphalt pavements by limiting permanent deformation (rutting), fatigue cracking, and low temperature cracking. This improvement happens by designating many rheological properties that are measured by equipment such as dynamic shear rheometer, bending beam rheometer, direct tension tester, rotational viscometers, pressure ageing vessel, and rolling thin oven film test [12].

This study investigates the effects of chemical composition on improving the rheological and physical behaviour of asphalt cement and recognizes it's the rheological properties of asphalt cement.

## 2. Materials Fractionation

In this work, a set of four asphalt cement with penetration grades of 20/30, 40/50, 60/70 and 85/100 was selected. Each asphalt cement was divided into four fractions following the Corbett method [13]. According to this method, the asphalt cement is separated into asphaltenes, resins, aromatics and saturates. The asphalt cement was first analysed by the asphaltene analyser to determine the asphaltene content. Samples were sufficiently dissolved in toluene before mixing with heated heptane. Then, the mixture was placed in the asphaltene analyser for measurement as shown in Fig. 1.

In the second step, the Fourier Transform InfraRed instrument (FTIR) was used to calculate the remaining fractions. Prior to analysis, the sample was sufficiently dissolved in toluene and then poured into the KBR plates and placed inside the device (Fig. 2). An infrared radiation is passed through the specimen and absorbed. The result was a fingerprint of the specimen called spectrum which shows the transmission and the absorption of the asphalt molecules. The results were compared with the infrared correlation chart.

## 3. Methods

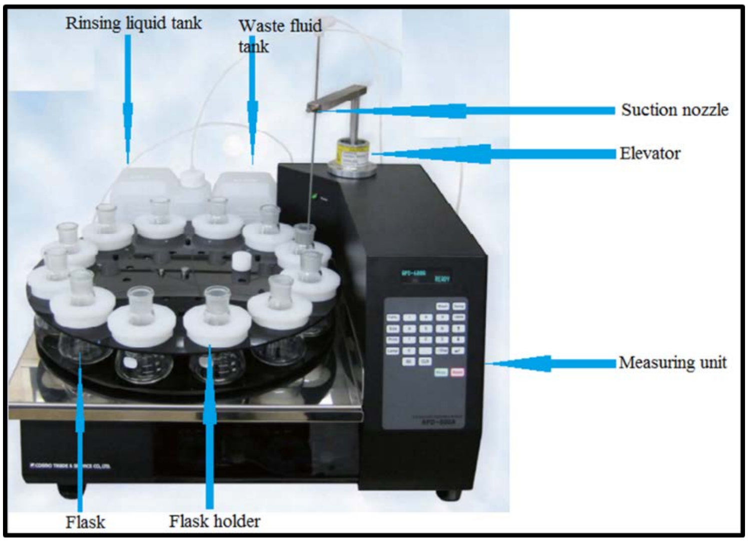
Dynamic Shear Rheometer and Rotation Viscometer were used to determine asphalt cement's rheological properties through penetration, softening point, and ductility.

### 3.1. Dynamic shear rheometer

A shearing force was applied on a thin sample of asphalt cement inserted between two plates; the upper plate was subjected to turning and the lower plate remained static as shown in Fig. 3. A 12.5 mm radius plate was employed for these tests with a sample thickness of 1 mm. DSR was used to determine the complex shear modulus $G^*$ and the phase angle $\delta$. The property of the asphalt cement at high temperature was determined, while the property of the asphalt cement at low temperature was not assessed because the fact that deformation for asphalt pavements in Iraq is due to high temperatures only. The rutting parameter $G^*/\sin\delta$, indicates the permanent deformation resistance of the asphalt cement. A high complex modulus and low phase angle benefit the high-temperature performance of the asphalt cement, which reduces the high-temperature flow deformation and increases the resistance to permanent deformation.

**(a) The asphalt content sample was dissolved by adding 1 ml of toluene.**

**(b) Adding 100 ml of heated Heptane to the sample.**

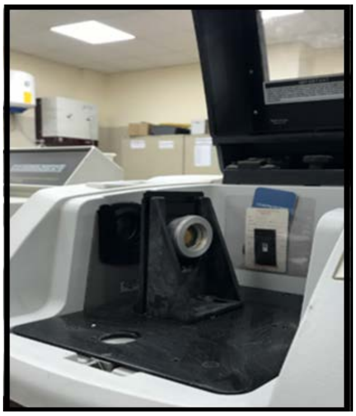



**(c) Asphaltene analyser device.**

**Fig. 1 Sample Preparation steps using asphaltene analyser.**

**(a) The sample poured into the cell.**

**(b) The sample is dissolved between the KBR plates.**



**(c) The sample inside the FTIR.**


**(d) The Asphalt spectrum.**

**Fig. 2. FTIR analysis of the Asphalt contents.**

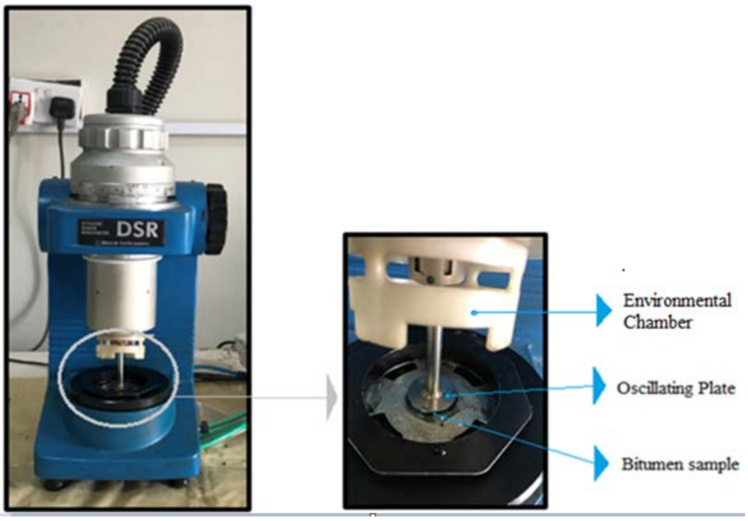



**Fig. 3. Dynamic shear rheometer (DSR).**

### 3.2. Rotational viscometer

This test was conducted for the asphalt cement at 135 °C and 165 °C, to assess temperatures for mixing and compaction during mix design. It is used to select the workability and the viscosity at high temperatures of asphalt cement. The rotational viscometer test for original asphalt cement can determine the compaction and blending temperatures of asphalt blends through the construction. The temperature was set at the desired value, the spindle (no.27) was dropped into the specimen chamber which contains 10.5 gm of asphalt cement.

### 3.3. Penetration test

The penetration test ASTM D5 [14] measured the consistency of asphalt cement, where a needle is applied to the specimen under fixed conditions of temperature, weight, and time. The penetration depth of the needle in the asphalt cement was measured in units of 0.1 mm.

### 3.4. Softening point test

According to ASTM D36 [15], the softening point test evaluates the behaviour of asphalt cement at increased temperatures. As the temperature is elevated, asphalt cement gradually changes from brittle to a soft and low viscous fluid. The test is done by pouring hot asphalt samples in closed rings and loading the samples by steel balls. The samples are placed in a beaker of cold water at a specified height above a metal plate. They are at a specified rate heated. As the temperature of water increase the temperature of asphalt also increased, by the weight of the steel ball the sample pulls down toward the plate. When the ball covered by sample touch the plate, the water temperature is measured and designated as the softening point of the asphalt.

### 3.5. Ductility test

According to ASTM D113 [16], ductility test measures the ability of asphalt cement to stretch. In other words, it is an indication of the asphalt cement to flow. A specimen of the asphalt cement was dragged apart at a standard rate at a fixed temperature (25°C), and the stretching before break estimated in units of cm.

## 4. Results and Discussion

Each type of asphalt cement was analysed to determine their chemical composition. The results showed that asphalt cement (20-30) has the highest percentage for asphaltene, resin and saturate but it has the lowest per cent for aromatic. On the other hand, asphalt cement (85-100) demonstrated the opposite. The chemical composition for the different asphalt types is shown in Fig. 4.

### 4.1. Effect of chemical composition on physical properties

The chemical composition embodied by (SARA fraction), Gastel index ($Ic$) and the ratio of the content of aromatics to asphaltenes ($Iar/as$) have an effect on physical properties represented by penetration, softening point, and ductility of asphalt cement as shown in Table 1.

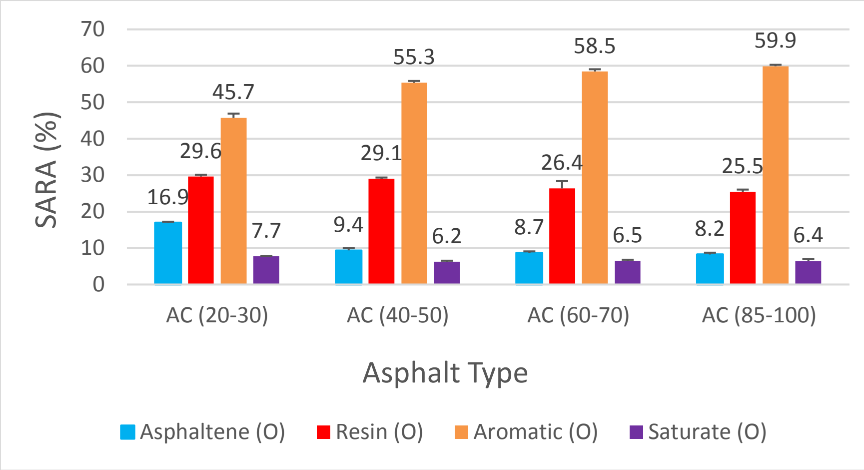



**Fig. 4. SARA Fractions.**

**Table 1. Chemical Composition and Physical Properties of Asphalt Cement.**

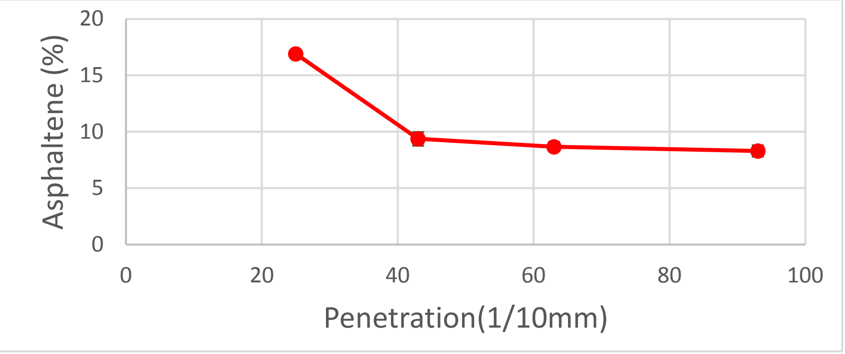
| Asphalt Type | Asphaltene Percentage | Gastel Index (*Ic*) | *Iar/as* | Penetration (1/10 mm) | (R&B) (°C) | Ductility (cm) |
|---|---|---|---|---|---|---|
| **AC (20-30)** | 16.9353 | 0.3276 | 2.6985 | 25 | 62 | 60 |
| **AC (40-50)** | 9.3688 | 0.1848 | 5.9071 | 43 | 55 | +158.5 |
| **AC (60-70)** | 8.6849 | 0.1790 | 6.7306 | 63 | 50 | +158.5 |
| **AC (85-100)** | 8.2320 | 0.1718 | 7.2743 | 93 | 48.5 | +158.5 |

### 4.1.1. Effect of chemical composition on penetration

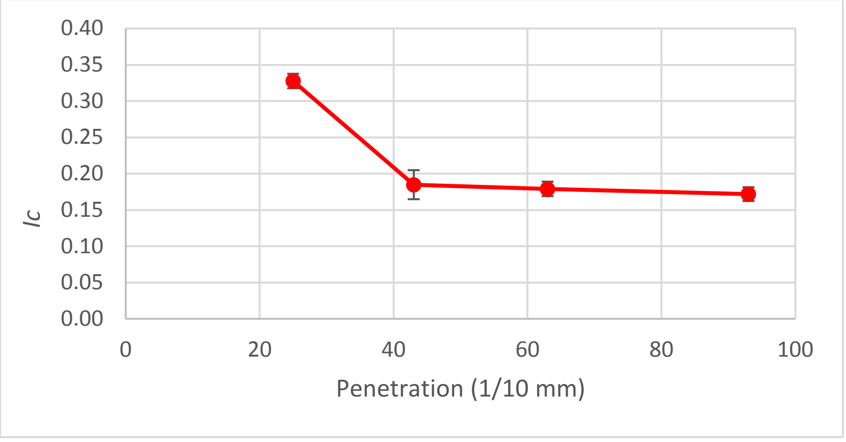
The percentage of asphaltenes affected the depth of the needle once it passed in the sample. Asphaltene content decreased by 44.67, 48.71, and 51.39% led to increasing penetration respectively by 72, 152, and 272 %, respectively. Decrease in Gastel index or colloidal index by 43.58, 45.36, and 47.55% led to increase the penetration by 72, 152, and 272 %, respectively. The ratio of aromatic to asphaltene has a positive relation with penetration. When *Iar/as* increases by 118.9, 149.42, and 169.56 % penetration increases with the same value as previously reported (Fig. 5). In other words, penetration is an indicator of the asphalt cement viscosity. The viscosity of asphalt cement decreases when the penetration increases due to one of these causes (decrease of Asphaltene content, the decrease in *Ic*, and the increase of *Iar/as*).

### 4.1.2. Effect of chemical composition on softening point

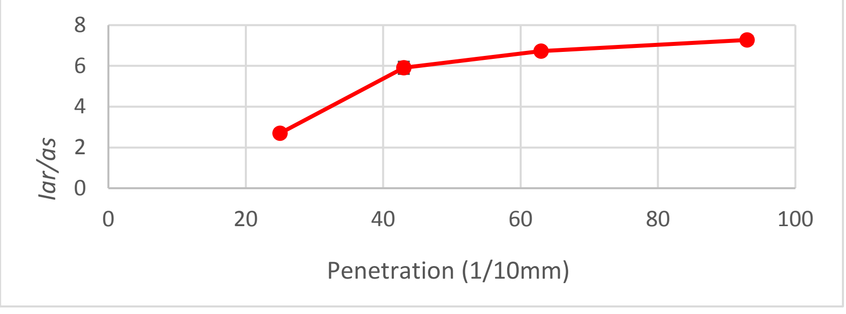
The relationship between the percentage of asphaltene and the softening point value demonstrated that an increase in asphaltene content by 5.5, 13.8, and 105.72% led to enhancing the softening point by 3.09, 13.4, and 27.83%, respectively. where, Fig. 6(a) showing that. A further correlation conducted between the Gastel index *Ic* and softening point (see Fig. 6(b)), when *Ic* increased by 4.19, 7.56, and 90.68% the softening point increased by 3.09, 13.4, and 27.835%, respectively. On the other hand, there is a negative relationship between softening point and *Iar/as* appeared, when *Iar/as* increased by 118.9, 149.42, and 169.56% softening point decreased by 11.29, 19.35, and 21.77%, respectively as shown in Fig. 6(c). Due to the positive relationship between the viscosity of the asphalt cement and the softening point, a rising asphaltene content, increasing Gastel index and decreasing *Iar/as* leads to an increase in the viscous of the asphalt cement.




**(a) The effect of Asphaltene percentage on penetration.**

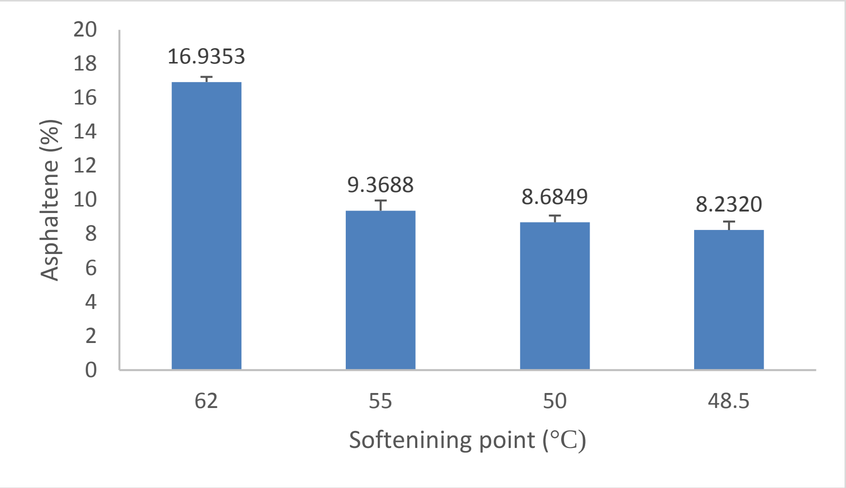



**(b) Relationship between Gastel index and penetration.**

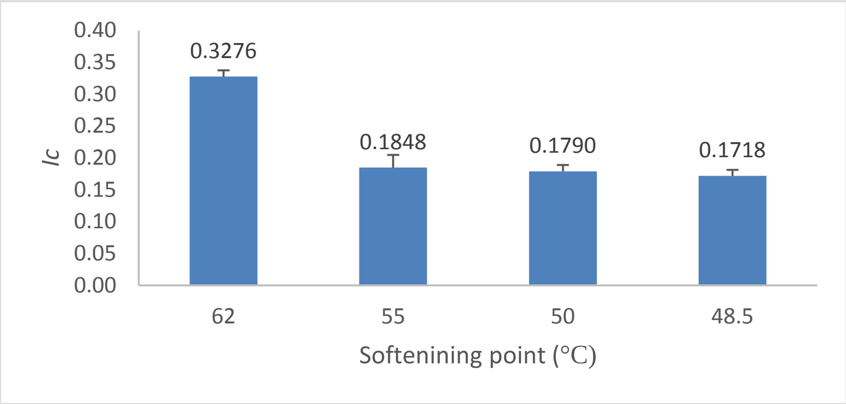



**(c) Relationship between *Iar/as* and Penetration.**

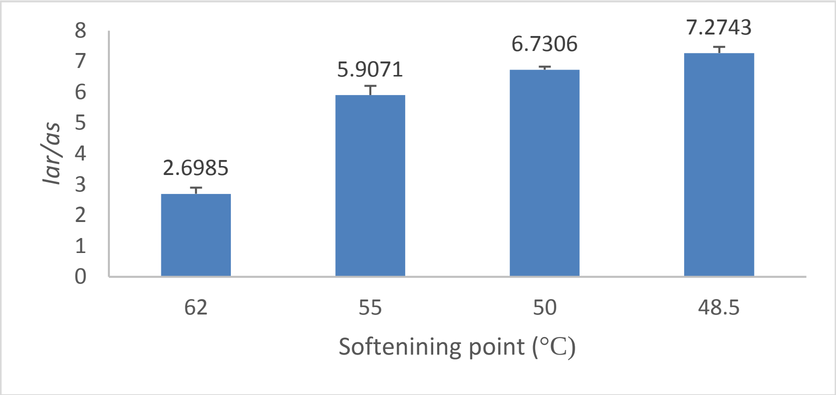
**Fig. 5. The Effect of Chemical Composition on Penetration.**




**(a) The effect of Asphaltene percentage on softening point.**



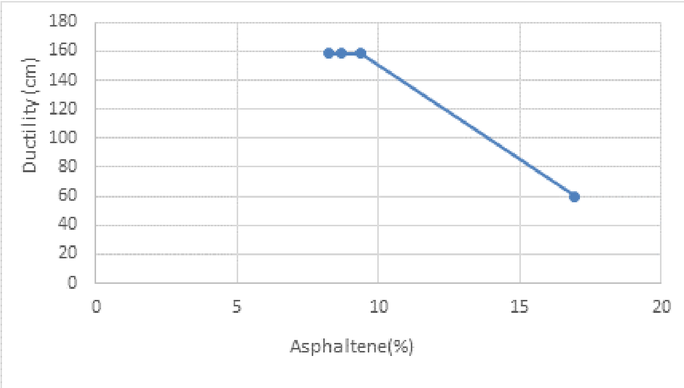

**(b) The effect of Gastel index on softening point.**



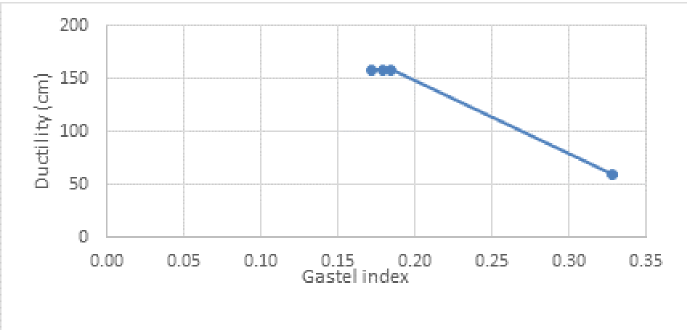

**(c) The effect of *Iar/as* on softening point.**
**Fig. 6. The effect of chemical composition on softening point.**

### 4.1.3. Effect of chemical composition on ductility

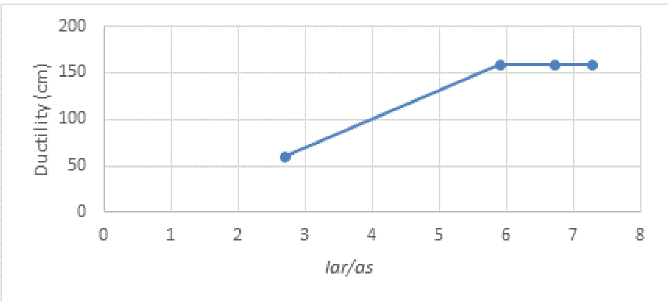
Ductility is not affected by little changes of chemical properties between samples. However, it is affected by substantial change, as shown in the change of asphalt type from AC (20-30) to AC (40-50) where the proportion of asphaltene and *Ic* reduced by 44.67, and 43.58%, while the ratio of aromatic to asphaltene increased by 118.9 %. These changes in the values led to a change ductility from 60 cm to more than 158.5cm, as shown in the Fig. 7. In other words, decreasing of asphaltene content, reducing in *Ic*, and the increase in *Iar/as* permits for asphalt cement to undergo great deformation or elongation.




**(a) The effect of Asphaltene percentage on ductility**




**(b) Relationship between *Ic* and ductility.**




**(c) Relationship between *Iar/as* and ductility.**

**Fig. 7. The effect of chemical composition on ductility.**

As a result, from the previous table and figures:

- Increase in asphaltene content led to the decrease in penetration, which is a measure of the consistency of asphalt cement (stiff or elastic). When the penetration decreases, it means that the asphalt became stiffer and demonstrated less elongation and flow, and it became more viscos as represented by softening point.
- The Gastel index demonstrates the colloidal system properties of the asphalt cement. A decreasing in Gastel index implies more peptized asphaltenes and a growing of sol character of the asphalt cement, leading to a decrease in viscosity and an increase in penetration and ductility.
- The aromatic content to asphaltene content ratio allows an estimation of the solvation capacity of the asphalt cement. An increase in this ratio (meaning aromatic content is larger than asphaltene content) implies more peptized asphaltenes and thus a decreasing in gel property of the asphalt cement. Consequently, it results in an increase in penetration and ductility and a decrease in viscosity represented by softening point.

## 4.2. Effect of chemical composition on rheological properties

Rheological properties (complex shear modulus $G^*$, phase angle $\delta$, $G^*/sin\delta$, performance grade PG and rotational viscosity) are affected by the change in chemical properties of asphalt cement as shown in Table 2 and Figs 8 to 12.

A change in asphaltene content led to a change in all rheological properties. The trend of the relation between asphaltene content and $G^*$ illustrate that the reduction in asphaltene content by 44.67, 48.71, and 51.39 % and result in a decrease of $G^*$ by 55.48, 75.26, and 79.52%, respectively. The complex shear modulus is a gauge of the stiffness of asphalt cement. Thus, downward asphaltene contents result in declined stiffness. The results show that $G^*/sin\ \delta$ and $G^*$ with asphaltene content have the same correlation, therefore, this similarity comes from $sin\delta$ for high $\delta$ almost equal to one. As explained, $G^*/sin\ \delta$ signifies rutting resistance, so a decrease in asphaltene content led to reduction of rutting resistance.

There is an inverse relationship exists between the phase angle and the asphaltene content. The viscoelastic deformation performance of asphalt cement is measured by phase angle. A phase angle of $\delta$ = 0° represents an ideal elastic performance, while a phase angle of $\delta$ = 90° represents an ideal viscous of the asphalt cement. Therefore, a high asphaltene content of asphalt cement results an increasing in elastic deformation performance. When asphaltene content increased by 5.5, 13.8, and 105.7% that led to a decrease in phase angle by 0.224, 0.449, and 18.876%, respectively.

Viscosity is a distinctive property of asphalt cement. It is the measure of the internal friction of fluid when a layer of this fluid is forced to move to another layer. Viscosity is a value which is highly dependent on temperature. Rotational viscosity was measured at different temperatures of 135°C and 165°C. As noted that the RV at 165 °C is less than RV at 135°C, which is reduced by the value of 2337.5 cP or 82.74% for asphalt AC (20-30), 362.5 cP or 74.35% for asphalt AC(40-50), 400 cP or 84.21% for asphalt AC (60-70) and 275 cP or 78.57% for asphalt AC(85-100). As previously explained, the relationship between heat and viscosity is inverse. Another relationship that is taken into a count is the relation between asphaltene content and rotation viscosity. While asphaltene content increased by 5.5, 13.8, and 105.7%, RVat135°C increased by 35.71, 39.28, and 707.14%, respectively, whereas RVat165 °C increased by 0, 66.66, and 550%, respectively.

Performance grade (PG) measures maximum and minimum pavement design temperature. In this study, only the maximum pavement design temperature was used. The trend of the relationship between asphaltene content and PG is directly. Increase of asphaltene content by 5.5, 13.8, and 105%, which led to increase of PG by 10.34, 10.34, and 41.37%, respectively. Thus, when asphaltene content increased, a maximum pavement design temperature increased as well.

**Table 2. Chemical composition and rheological properties.**

| Asphalt Type | AC (20-30) | AC (40-50) | AC (60-70) | AC (85-100) |
|---|---|---|---|---|
| ***G****(kPa) | 4.65 | 2.07 | 1.15 | 0.952 |
| ***δ*** | 72.2 | 88.6 | 88.8 | 89.0 |
| ***G*/sinδ***(kPa) | 4.89 | 2.07 | 1.15 | 0.952 |
| **PG** | 82 | 64 | 64 | 58 |
| **RV at 135 °C** | 2825 | 487.5 | 475 | 350 |
| **RV at 165 °C** | 487.5 | 125 | 75 | 75 |
| **Asphaltene** | 16.9353 | 9.3688 | 8.6849 | 8.2320 |
| ***Ic*** | 0.3276 | 0.1848 | 0.1790 | 0.1718 |
| ***Iar/as*** | 2.6985 | 5.9071 | 6.7306 | 7.2743 |

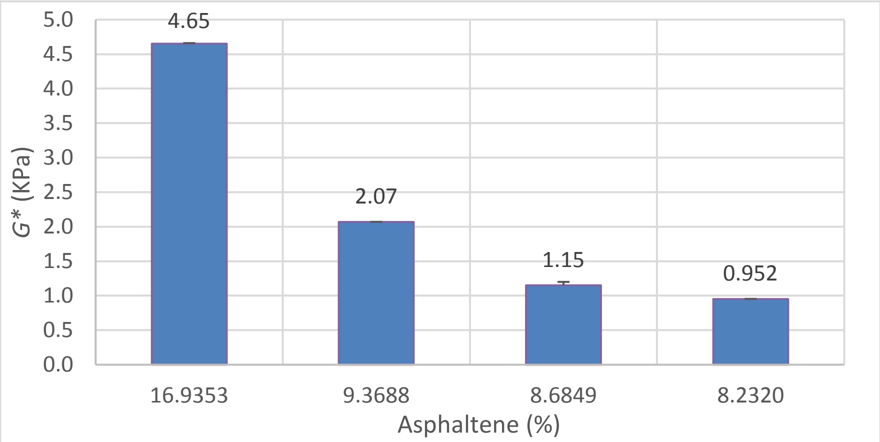



**Fig. 8. The effect of Asphaltene on *G**.**

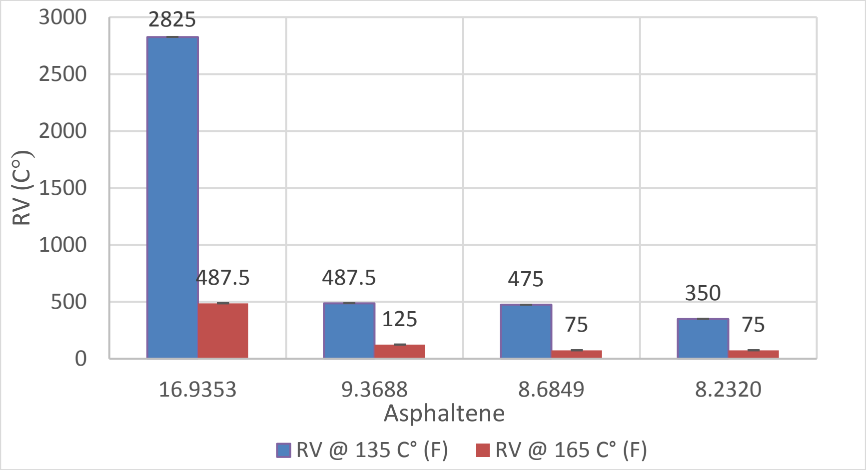



**Fig. 9. The effect of Asphaltene on RV.**

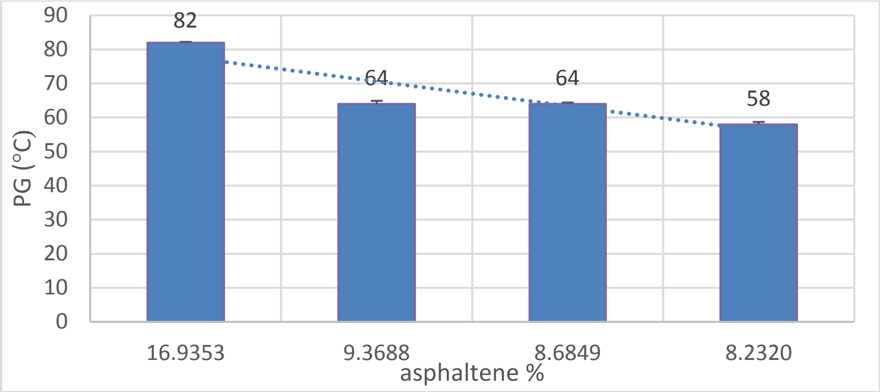



**Fig. 10. The effect of Asphaltene on PG.**

The relationship between complex shear modulus and Gastel index is positive. When *Ic* decreased by 43.59%, 45.37% and 47.56%, it led to a decrease in *G** by 55.48, 75.26, and 79.52%, respectively. Therefore, when gel characteristic decreases, the stiffness of asphalt cement also decreases.

When *Ic* decreases by the same percentage as explained above, $G^*/\sin\delta$ decreased by 57.66, 76.48, and 80.53% respectively. So, the decreases in gel character of asphalt cement lead to the decrease in a rutting resistance.

Phase angle with the Gastel index shows negative relationship. When Gastel index increased by 4.19, 7.56, and 90.68%, that produced a decrease in phase angle by 0.224, 0.449, and 18.87% respectively. Therefore, the increase in gel characteristic of asphalt cement led to the decrease in viscoelastic properties.

There is a direct relationship between Gastel index and performance grade. When *Ic* increased by 4.19, 7.56, and 90.68% respectively, PG increased by 10.34,10.34, and 41.37. Increasing asphalt gel properties led to an increase in the maximum pavement design temperature.

The relationship between viscosity and Ic was positive. When the Gastel index increased by 4.19,7.56, and 90.68%, RVat135 °C increased by 35.71 ,39.28, and 707.14%, respectively. While RVat165 °C increased by 0, 66.66, and 550%, respectively. Therefore, an increase in the gel properties of asphalt cement led to an increase in viscosity.

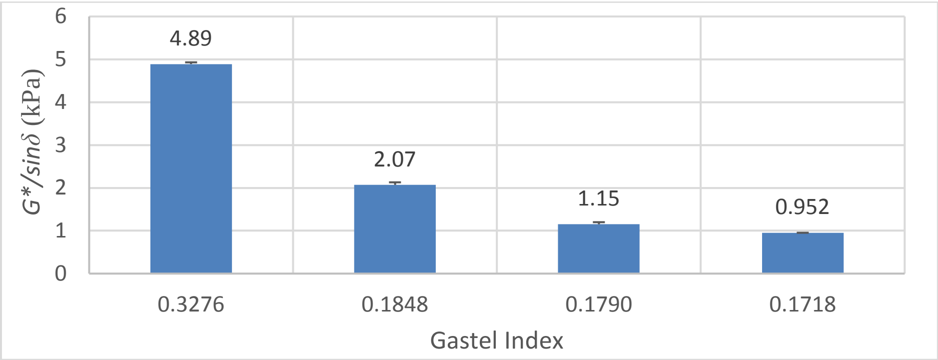



**Fig. 11. The Effect of *Ic* on *G*/sinδ*.**

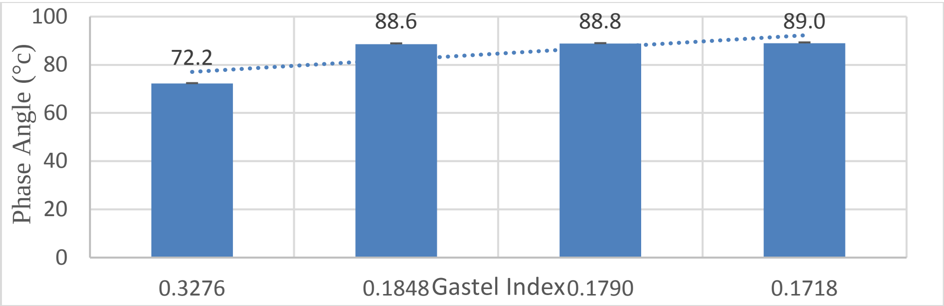



**Fig. 12. The Effect of *Ic* on *δ*.**

The aromatic content to asphaltene content ratio illustrates an estimation of the solvation capacity of the asphalt cement. The increase in aromatic content to asphaltene content ratio means increase peptise of the asphaltene and so a reduction in gel property of the asphalt cement. There is an inverse relation appeared between RV and *Iar/as*. Increasing the *Iar/as* by 118.9, 149.42, and 169.59% led to a decrease of RV at135°C by 82.74, 83.18%, 87.6%, respectively, while the decrease in RVat165°C by 74.35, 84.61, and 84.61%, respectively.

Another inverse relationship was observed between *Iar/as* and performance grade. *Iar/as* increased with the same values as shown in the previous paragraph, resulting in a decrease of PG by 21.95, 21.95, and 29.26%, respectively.

An increase in *Iar/as* by 118.90, 149.42, and 169.59% led to an increase in phase angle by 22.71, 22.99, and 23.26%, respectively. Due to this relationship, an increase of aromatics content to asphaltenes content ratio resulted in a decrease in gel property of the asphalt cement which in turns raising the phase angle (viscous behaviour of the asphalt cement).

An increase in the ratio of aromatic to asphaltene by 118.9, 149.42, and 169.59% led to a decrease in $G^*/sin\delta$ by 57.66, 76.48, and 80.53%, respectively while the decrease of $G^*$ by 55.48, 75.26, and 79.52%, respectively. The stiffness and rutting resistance of asphalt cement was reduced by the increase of *Iar/as* (reducing the gel character of the asphalt cement).

## 5. Conclusions

In this research, the chemical compositions of the asphalt cement have been shown to have an influence on the physical and rheological properties. The asphalt cement gel property (represents the chemical properties) increases due to one or more of the following reasons:

- Decrease of the ratio of aromatic to asphaltene content.
- Increase of the Gastel index.
- Enhance the asphaltene content.

It was noted that the increase in the gel characteristics of the asphalt cement leads to changes in physical and rheological properties, as discussed below:

- Increasing viscosity is illustrated by the value of softening point and the rotational viscosity.
- Increasing stiffness is illustrated by the complex shear modulus and Penetration.
- Less elongation and flow are captured by ductility.
- An increasing elastic deformation property is illustrated by the phase angle $\delta$.
- Increase in rutting resistance is showed by the complex shear modulus over sin delta ($G^*/sin\delta$).
- Increase pavement design temperature is showed by the performance grade.

## Disclosure Statement

The authors declare no financial or commercial competing interests.

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**Nomenclatures**

| | |
|---|---|
| *G** | Complex shear modulus (kPa) |
| *Ic* | Gastel index |
| *Iar/as* | The ratio of aromatic content to asphaltene content |

***Greek Symbols***

| | |
|---|---|
| *Δ* | Phase angle |
| **Abbreviations** | |
| AC | Asphalt Cement |
| ASTM | American Society for Testing and Materials |
| DSR | Dynamic Shear Rheometer |
| PG | Performance Grade |
| RV | Rotation Viscosity |
| SARA | Saturate, Aromatic, Resin, Asphaltene |

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