Rheological Modification for Asphaltic Materials Using a Mixture (Polycarbonate: Poly Methyl Methacrylate) and Microwave Radiation

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ABSTRACT

Obtaining asphalt that suits the nature of the countries it is used in, is of great importance. Asphalt constitutes an important component in the paving of roads, mastic and flattening substance. This study was included the modification of the asphaltic material by the use of a mixture of polycarbonate and polymethyl methacrylate after crushed to obtain low molecular weight. The study included two paths: In the first asphaltic materials was treated with different percentages of a mixture in the presence of anhydrous aluminium chloride (0.06% of weight) as a catalyst for this reaction. The mixture was heated at 150°C for 30 minutes. In the second path, the process of mixing was performed with the existence of a 360 watt microwave radiation for 15 minutes. The process of mixing was repeated in the presence of sulphur (1% of weight) instead of anhydrous aluminium chloride and under the same conditions. Properties of the original and modified asphalts were determined, and this included the measurement of ductility, penetration, softening point, penetration index and calculating the percentage of separated asphaltenes. Additionally, Marshal test, chemical Immersion test and aging were measured for some of the samples. Good samples that can suit Iraq’s climate were obtained from this study.

INTRODUCTION

Asphalt is a hydrocarbon substance that varies between semi-solid to the liquid state of high viscosity, of dark brown color and a “sticky” nature. It is a hydrocarbon compound produced as a residual of raw oil distillation (Parkash, 2010) Asphalt has many names; in Europe the term (Bitumen) is used to refer to asphalt which is the name used in North America; outside North America, the term (asphalt) is used to signify the combinations of bitumen with metals (Zhang and Greenfield, 2008) Many studies have been performed on asphalt considering the diversity of its natural and industrial resources and as a result to its use basically in paving roads and other various uses due to its good adherence with different solid substances, as well as its suitable viscosity let alone its availability in respectively low prices. These studies showed that its properties such as viscosity and thermal sensitivity can be enhanced by using different mixtures or by chemical modification. By resorting to previous researches and studies, we find many researches and different methods used in modifying the rheological properties of asphalt such as (Habib et al., 2011) studied the rheological properties of asphalt using...
polyethylene and polypropylene, the study showed an enhancement in the rheological properties and that the effect of these polymers was in penetration more than in softening point. The best samples were obtained at a percentage of less than (3%). (Wang et al., 2011) used microwave to heat asphalt with carbonyl ferrous compounds. The results showed that the heat produced by the microwave helped enhancing the properties of asphalt mixtures such as dynamic strength, stability and flow for the marsh-al test. (Djaffar et al., 2013) managed to study the rheological and stability properties during storage of modified asphalt by adding the polymer styrene-ethylene- butylene- styrene (SEBS). The study confirmed the stability of the rheological properties (ductility, softening point, penetration index, temperature sensitivity) of modified asphalt through their measurement after storage. (Shirini and Imaninasab, 2016) studied the rheological properties of the porous asphalt modified with rubber and compared it with mixtures of porous asphalt modified by (SBS), and they found that adding 10% of rubber fragments led to enhancing the performance of porous asphalt in resisting moulding. (Tang et al., 2018) studied the rheological properties of asphalt modified with polymer and sulphur. In this study hybrid asphalt bonds which contain fragments of rubber and styrene-butadiene- (SBS) styrene and sulphur were prepared in different percentages, and then a test was performed to check the stability of the storage, the performance classification (Superpave), the stability and the flow.

The results indicated that the use of (SBS) with sulphur help enhancing the properties of high temperature and the elastic behaviour of the asphalt bond. The study recommended the use of sulphur (0.2% - 0.3 %) of weight. In another study, (Lin et al., 2019) managed to enhance the properties of asphalt using garbage sacks which are hard to recycle. The results of this modification showed increased viscosity and reduced penetration and softness, the study asserted that the modification was a physical one. (Khiavi et al., 2020) managed to modify the rheological properties of asphalt at high temperatures through adding hydroxy ethyl methyl acrylate (PHEMA) by different percentages. The samples were homogenous mixtures. The results showed an enhancement in permanent deformation resistance of asphalt in high temperatures. Additionally, many tests were performed like light scanning, thin tissues furnace, permeability, softness and viscosity, and the results were good. There are a significantly large number of studies in this regard; accordingly, we modified the Iraqi asphalt with waste polymer substances where results that suit the Iraqi weather conditions were obtained.

**Experimental**

A certain weight of asphalt was taken and put into the device of processing asphaltic substances. The sample was treated with different percentages of a mixture (poly carbonate- Poly methyl methacrylate). 0.06% of weight anhydrous aluminium chloride was added to the mixture as a catalyst for this reaction. The mixture was heated to 150 c for 30 minutes. The samples were subjected to microwave ray of 360 watt power, for time (15) minutes.

The rheological properties for the original and the modified samples were calculated, and this included the measurement of ductility (ASTM D113-99, 2017), penetration (ASTM D5-97, 1998), softening point (ASTM D36-95, 2000) and calculating the penetration index (Yoder and Witczak, 1964). Also, calculations were made to check the separated percentage of the asphaltens (ASTM D6560-17, 2017), the Marshal test (ASTM D1559-89, 2014), aging test (ASTM D1754-97R, 2002) and the chemical immersion (Speight, 2015) for some samples.

**RESULTS AND DISCUSSION**

Obtaining asphalt of rheological properties that suit the climate of its country is of high importance. Therefore, researchers were keen on making componential modifications by way of deforming the original asphalt structure. Recently, Polymeric wastes are used in most of the modifications.

![Figure 1: The relation between the percentages of the additive and the rheological properties of asphalt](attachment:image)

Our study focused on the use of crushed plastic wastes manufactured from polycarbonate and poly methyl methacrylate.

The study included processing asphaltic materials with a polymer mixture of the above mentioned with a ratio of (1:1). The mixture was added in different percentages that ranged between (0.5% - 4% of weight). The process was performed with the use...
Table 1: The Rheological properties of asphalt modified with different percentages of the additive.

| Sample no | Polycarbonate: poly methyl methacrylate mixture | Ductility cm. 25°C | Penetration 100gm. 5sec. 25°C | Softening point 25°C | Penetration index (PI) | Asphaltenes % |
|-----------|-------------------------------------------------|-------------------|-------------------------------|---------------------|-----------------------|--------------|
| As0       | 0                                               | 100⁺              | 44.6                          | 50                  | -1.448                | 17.81        |
| As1       | 0.5                                             | 100⁺              | 44.7                          | 58.7                | 0.387                 | 18.21        |
| As2       | 1                                               | 100⁺              | 41.8                          | 59                  | 0.398                 | 18.58        |
| As3       | 2                                               | 100⁺              | 40.6                          | 61                  | 0.721                 | 20.82        |
| As4       | 3                                               | 85                | 39.5                          | 62.7                | 0.976                 | 21.61        |
| As5       | 4                                               | 73                | 38.1                          | 64                  | 1.127                 | 21.48        |

Table 2: The rheological properties of asphalt modified with different percentages of the additive in the presence of sulfur.

| Sample no | Polycarbonate: poly methyl methacrylate mixture | Ductility cm.25°C | Penetration 100gm. 5sec. 25°C | Softening point 25°C | Penetration index (PI) | Asphaltenes % |
|-----------|-------------------------------------------------|-------------------|-------------------------------|---------------------|-----------------------|--------------|
| As0       | 0                                               | 100⁺              | 44.6                          | 50                  | -1.448                | 17.81        |
| As6       | 0.5                                             | 100⁺              | 42.8                          | 56                  | 0.384                 | 19.61        |
| As7       | 1                                               | 100⁺              | 41.2                          | 58                  | 0.159                 | 20.38        |
| As8       | 2                                               | 100⁺              | 41.7                          | 57                  | 0.013-                | 21.85        |
| As9       | 3                                               | 100⁺              | 40.8                          | 59.5                | 0.442                 | 21.89        |
| As10      | 4                                               | 88                | 39.1                          | 61.7                | 0.768                 | 23.62        |

Table 3: Marshall Test for the original asphalt and the modified asphalt

| No. of asphalt sample | Percentage asphalt added to aggregates % | Stability (KN) | Flow (mm) | MQ |
|-----------------------|------------------------------------------|----------------|-----------|----|
| As0                   |                                          | 11.3           | 5.1       | 2.21 |
| As1                   | 4.5%                                     | 15.8           | 3.5       | 5.28 |
| As6                   |                                          | 16.1           | 2.5       | 6.88 |
| As*                   | 3%-5.5                                   | 7 Minimum      | 4-2       | 3-5 Minimum |

As * specifications of the Iraqi roads and bridges, Authority.

Table 4: Chemical Immersion results

| No. of modified sample | Percentage of Na₂Co₃gm | R&WNO R&WNO For the original asphalt | R&WNO R&WNO For the modified samples |
|------------------------|-------------------------|--------------------------------------|--------------------------------------|
| 0.025                  | 1                       |                                      |                                      |
| 0.041                  | 2                       |                                      |                                      |
| 0.082                  | 3                       | 3                                    | 3                                    |
| 0.164                  | 4                       |                                      |                                      |
| 0.328                  | 5                       |                                      |                                      |
| 0.656                  | 6                       |                                      | 6                                    |
| 1.312                  | 7                       |                                      | 7                                    |
| 2.624                  | 8                       |                                      |                                      |
| 5.248                  | 9                       |                                      |                                      |
of anhydrous aluminium chloride (0.06% of weight) as a catalyst for the reactions. Table 1 and Figure 1. Show the most important results obtained this treatment will be discussed.

From Table 1 it is clear that adding polymer mixture to asphalt led to obtaining asphalt samples of good rheological properties up to 2%; the ductility values of all the samples were more than 100, and the penetration values were within the accepted range for the paving asphalt demonstrated in Tables 6, 7, and 8. (Technical specification for civil work, 2019), Table 7 (ASTM D491-88, 2006) and Table 8. Sample As 1 was taken as the best sample to be used in later measurements.

Adding polymer to asphaltic materials is a very sophisticated process; the main reaction that takes place during mixing the additive to the asphaltic or petroleum materials, in general is a reaction of (alkylation reaction). Particles of crushed polymer bind to asphalt with the existence of anhydrous aluminium chloride as a catalyst for the reaction. This reaction leads to an increase in the molecular weight of the processed asphalt. This increasing of molecular weight was apparent in the increase of the percentages of the isolated asphaltenes with the increase of the added polymer.

### Table 5: Aging results for the parent and modified samples

| Sample no. | Rheological properties | Before test | After test |
|------------|------------------------|-------------|------------|
| As0        | Softening point (°C)   | 50          | 53         |
|            | penetration (100gm.5sec.25°C) | 44.6       | 41.1       |
|            | ductility cm. 25°C     | 100⁺        | 100⁺       |
|            | Weight loss %          | ---         | 0.05       |
| As1        | Softening point (°C)   | 58.7        | 61         |
|            | penetration (100gm.5sec.25°C) | 42.7       | 41.1       |
|            | ductility cm. 25°C     | 100⁺        | 100⁺       |
|            | Weight loss %          | ---         | 0.027      |
| As6        | Softening point (°C)   | 55          | 58.5       |
|            | penetration (100gm.5sec.25°C) | 42.8       | 40.7       |
|            | ductility cm. 25°C     | 100⁺        | 100⁺       |
|            | Weight loss %          | ---         | 0.031      |

### Table 6: Iraqi paving asphalt

| Rheological properties | Minimum | Maximum |
|------------------------|---------|---------|
| Softening point (°C)   | 54      | 60      |
| Penetration (100gm.5sec.25°C) | 40       | 50      |
| Ductility (cm.25°C)    | 100     | -       |

### Table 7: Flattening asphalt properties

| Rheological properties | Minimum | Maximum |
|------------------------|---------|---------|
| Softening point (°C)   | 57      | 66      |
| Penetration (100gm.5sec.25°C) | 18       | 40      |
| Ductility (cm.25°C)    | 10      | -       |

### Table 8: American mastic properties

| Rheological properties | Minimum | Maximum |
|------------------------|---------|---------|
| Softening point (°C)   | 54      | 65      |
| Penetration (100gm.5sec.25°C) | 20       | 40      |
| Ductility (cm.25°C)    | 15      | -       |
In an attempt to enhance the properties of asphalt more, asphalt was processed with the same mixture in the same circumstances of the first reaction such as the intensity of radiation and the time of reaction, but we skipped the use of anhydrous aluminium chloride and replaced it with sulfur (1% of weight). Table 2 and Figure 2 show the most important results obtained this treatment will be discussed.

Sulfur changes at temperature of 150°C to its free radical i.e. mixing the polymer additive with asphalt at 150°C leads to changing sulfur to its free root.

Moving to the second step which is the treatment with microwave radiation, the reaction of the polymer with asphalt completes and sulfur forms sulfurous bridges that give the asphalt sample its desired elasticity (Tang et al., 2018).

Looking at the Table 2, we find that ductility values were good to excellent at all percentages. Going back to the comparison between the results obtained and the standard specifications given in Tables 6, 7 and 8, the choice was set on sample As1& As6 as the best sample for the upcoming measurements.

In order to know the stability and Flow value, Marshall test was performed on the original sample As0, As1 and As6 that represent the best-obtained results. Table 3 show the most important results obtained this treatment will be discussed.

From the table, it is clear that the obtained values of the modified asphalt are better than the original sample. The results in the table show that pavement that used the modified asphalt is more resistant to deformation. Deformation of asphaltic roads is a result of being subjected to recurrent loads of different transportation means. MQ value which was higher in the modified asphalt gives an evidence to that modified asphalt is more resistant to permanent deformation than the original asphalt. Table 4 show the chemical Immersion for the original asphalt, and the best samples were obtained.

The chemical Immersion test is a measurement of the adhesion ability of asphalt to aggregate. From that table, it is clear that the modified samples were better than the original in their ability to adhere to aggregate.

Additionally, aging was measured for the original sample and for the best modified samples, Table 5 demonstrates the obtained results.

Based on all what was mentioned above, it is clear that the process of modification with the use of the suggested a mixture in the study gave good results. It also gave asphalt samples suitable for use in the climate of Iraq.

CONCLUSIONS

After completing this study, a number of conclusions were reached, the most important of which can be included in the following points:

1. The use of polymeric mixtures gave a better results than if each polymeric additive was used only.
2. The use of microwave radiation in the modification of the rheological properties of asphalt to alter the rheological, reducing the time needed to complete the reaction of modification.
3. From this study, we obtained some samples with rheological properties suitable for paving operations in Iraq.
4. The use of polymeric wastes in modification processes led to the occurrence of alkylation reaction between both the additive and the asphalt in the presence of anhydrous aluminium chloride as a catalyst for this reaction.

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Conflict of interest
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