Evaluating Modified Asphalt Binder Comprising Waste Paper Fiber and Recycled Low-Density Polyethylene

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Abstract: The main concept of using waste and recycled materials as modifiers for construction materials, is to improve their performance, keep the resources, and conserve the environment from pollution problems. This investigation is tended to evaluate the effect of using two different types of modifiers on the physical properties of asphalt binder. The modifiers are Recycled-Low-Density-Polyethylene (R-LDPE) polymer and waste-Paper Fiber (w-PF), they were mixed individually and collectively with neat asphalt binder. The first set of modified binders comprised neat binder with 3% R-LDPE alone, the second is comprised 0.3%, 0.5% and 0.7% of w-PF alone, while the third comprised collective modifiers of 3% R-LDPE and 0.3%, 0.5% and 0.7% w-PF. The influence of these modifiers on neat asphalt binder was evaluated in terms of penetration (Pen.), softening point (SP), penetration index (PI), ductility, rotational viscosity (RV), penetration aging index (PAI) and softening point index (SPI). The obtained results indicate that the utilization of modifiers helps in improving the physical properties of asphalt binders by different levels. The modification by collective modifiers leads to an increase SP, RV, PI, PAI, and SPI to 65%, 2 times, 1.044, 0.93, 2.5 respectively compared to neat bitumen. As well, leads to a decrease in both Pen. and ductility to the levels 45% and 79% respectively. This indicates that the use of waste and recycled modifiers especially in the combined form encourage the production of comparative modified asphalt binder.

Keywords: Bituminous binder: mechanical properties; modified asphalt binder; recycled low-density polyethylene; recycled materials; waste paper.

1. Introduction
The use of recycled and waste materials in construction works is rising nowadays due to the higher amounts of wastes that are produced from the continuous production processes and modern lifestyle
[1]. That makes the possibility of dump these wastes is difficult and needed high costs because it primarily depends upon landfilling to eliminate their effects and problems [2, 3]. Therefore, the incorporation of these waste materials as modifiers into construction materials can contribute to reducing the landfilling costs of these wastes, then conserve the surrounding environment from pollutants [4-7]. Furthermore, that can be helpful in the reduction of construction costs and enhance the performance of construction materials [8].

From this point of view, many asphalt researchers around the world are tended to use different modifiers with asphalt binders to improve their properties and performance. Asphalt binder represents a sensitive material that characterizes by its visco-elastic properties, it is more susceptible to aging, and its mechanical response dependent on the loading time and temperature [9]. This action may be led to an increase in the appearance of more potential failures like fatigue and rutting under repeated traffic loads [10, 11]. Therefore, the utilization of the various modifiers i.e., virgin polymers, fibers and ashes (including the recycled) can help to enhance its properties, as well as, increase pavement service life. The more common recycled materials used recently are waste polyethylene polymers (like Low-Density-Polyethylene -LDPE, Linear Low Density Polyethylene -LLDPE, High Density Polyethylene -HDPE, polypropylene and their co-polymers), bio-oils derived from waste wood [12], crumb rubber from waste tire [13], municipal solid waste incinerator ash [14], different types of fly ashes and waste materials [15, 16].

At this point, a number of researchers such as Polacco et al. [17], Behnood et al. [18], Al-Busultan et al. [7], and others tend to investigate the effect of recycled polymers like low-density polyethylene (R-LDP), on the properties of asphalt binder and, as well as, mixture. They found that the use of such modifiers has a significant effect on the properties of the asphalt binder itself. Where R-LDP helps in a decrease both penetration and ductility of asphalt and increase its softening point and viscosity to a specified level of modifier content. This also reflected positively on the mixture performance. That behavior returns to the nature of R-LDP that represents one of the polymerization products formed from one monomer. This in turn creates a polymer network interlock between asphalt molecules during a series of chemical reactions [19], and finally helps enhance the asphalt mixture flexibility and as well its durability. Liang et al. [20] also studied the effect of different polyethylene types such as LDPE, HDPE, and LLDPE polymers on the rheological behavior of asphalt. they found that the use of these polymers helps in enhancing the resistance of the asphalt to rutting, cracking and low temperature cracking.

On the other side, other researchers studied the effect of comprising fibers with asphalt to improve its properties like, Afonso et al. [21] who studied the effect of cellulose fibers and Chew et al. [22] who studied the effect of paper mill sludge, etc. Generally, they found that the fiber properties (i.e., length, adhesion, absorption and swelling) play an important role in forming the three-dimensional network into asphalt molecule, and then works on bond these molecules with each other, as confirmed previously by others [23]. That in turn results in increased the interlock between the asphalt binder and aggregate particles, then, the performance of mixture to moisture, fatigue, thermal cracking and rutting are improved [24, 25], as well as, helps in eliminating the separation phenomena of binder from aggregate [26, 27].

It is worth mentioning that surveying the literature disclosed the limited studies that use collective sustainable materials as binder modifiers. Therefore, in order to achieve the sustainability principle, this research investigates the incorporation of two modifiers with asphalt binder: waste-Paper Fiber (w-PF) (with three contents 0.3%, 0.5% and 0.7%), and Recycled-Low-Density-Polyethylene (R-LDPE) polymer with one content equal to 3%. Due to the widespread of these materials as a source of the wastes of both papers and bags of domestic goods respectively. The effect of them was studied in terms of Penetration (Pen.), Softening Point (SP), ductility, Rotational Viscosity (RV) and aging of the asphalt before and after modification.
2. Materials

2.1. Bitumen

Neat Bitumen (B0) with 40-60 penetration grade supplied from a local refinery in Nasiriyah was adopted in this research, the physical properties of this binder are summarized in Table 1. The neat asphalt binder was inspected to confirm the requirement of the General specification of roads and bridges (GSRB) specification [28].

| Property                                | Measured values | ASTM specification |
|-----------------------------------------|-----------------|--------------------|
| Penetration (25 °C, 0.1 mm, 5 sec, 100 g) | 42.8            | D 5-5a [29]        |
| Softening point (R&B), °C               | 44              | D 36-95 [30]       |
| Ductility at 25 °C, cm                  | 143             | D 113-99 [31]      |
| Penetration index (PI)                  | -2.603          | [32]               |
| Rotational Viscosity @135 °C, Pa. s     | 0.86            | ASTM D4402 [33]    |
| Flash point, °C                         | 355             | ASTM D92 [34]      |
| Solubility, %                           | 99              | ASTM D2042 [35]    |

2.1.1. Recycled-Low-Density Polyethylene (R-LDPE)

The used R-LDPE modifier in this research is a powder recycled material supplied from a small recycling factory located in Karbala City. The physical properties of this modifier are presented in Table 2, and its appearance appears in Figure 1.

| Properties                                | Amount |
|-------------------------------------------|--------|
| Density, g/cm³                            | 0.91   |
| Tensile strength, MPa                     | 8.5    |
| Tensile elongation, %                     | >350   |
| Melting temperature, °C                   | 110- 120 |
| Flexural modulus, MPa                     | 7.2    |
| Hardness shore D                          | 45     |

2.1.2. Waste-Paper Fiber (W-PF)

W-PF is a type of cellulose fibers materials that are produced from the cut-off of the waste papers into small pieces as presented in Figure 2. The chemical components of this material are similar to that for Portland Cement namely SiO₂, Al₂O₃, K₂O, CaO. Where the presence of calcite in its chemical composition provides good adhesion between aggregate and asphalt [22].
3. Experimental work

3.1. Preparation of Modified Binder

In this research, asphalt binder was modified in two different cases, the first by comprising R-LDPE or w-PF in a single form with the binder, while the second by comprising both modifiers. A shear mixer device with a rate of 3000 rpm was used to prepare a modified binder for one hour. Three dosages from w-PF (i.e., 0.3, 0.5%, and 0.7% by total weight of binder) were suggested by authors, while one dosage of R-LDPE (i.e., 3%) by total weight of binder was adopted, as proved by other research studies as a significant dosage [36-38]. However, the collective blends of modifiers (MC) comprising 3.3%, 3.5% and 3.7% as a part of binder weight.

The preparation is initially started by heating the neat asphalt binder in an oven at 170 °C for more than one hour to make it fluid enough for mixing. A specific experimental procedure was used to obtain a homogeneous bitumen-modified blend. After that, the fluid binder was placed into the shear mixer, then the modifier materials were added slowly to the heated bitumen, while the mixing continued at a constant speed rate to prevent the agglomeration of these modifiers. This procedure was repeated for each additive content until finished all types of the modified binder [39, 40].

3.2. Asphalt characterizing indices

The physical properties of the neat and modified binder were evaluated using a set of three tests: Penetration (Pen.), Softening Point (SP) and ductility depending on the procedures recommended by ASTM D5-D5M [29], ASTM D36 [30] and ASTM D113 [41], respectively. Moreover, the index of bitumen sensitivity to temperature was determined depending on the results of penetration and softening point, and by using Equation (1) as reported by Read and Whiteoak [32], Jun et al.[42] and Ameri et al. [11].

\[
PI = \frac{1952 - 500 \log pen - 20SP}{50 \log pen - SP - 120} \quad (1)
\]

Where: \( PI \) is penetration index, \( Pen. \) is the depth of penetration at 25 °C and \( SP \) is softening point temperature.

Another physical test was conducted to describe the rheological properties of the binder is the Rotational Viscosity (RV). The test was done at elevated temperatures ranged from 135 °C to 200 °C.
to display the effect of variation in temperature on bitumen fluidity. The procedure recommended by ASTM D4402 [33] was followed. The rotational viscosity criterion obtained from this test represents a function of the torsion required to measure the relative rotational speed of the spindle that is lowered into a bitumen sample.

Moreover, the effect of short-term aging on the properties of the binder was investigated, depending on the thin film oven test (TFOT) procedure recommended by ASTM D1754 [43]. That requires subjecting the bitumen sample to 163 °C for five-hours. Then the amount of the effect of temperature determined as a function of the aging index of physical properties of bitumen before and after subjected to short-term aging. Equations 2 and 3 were adopted to calculate the amount of loss in bitumen in terms of penetration and softening point before and after aging, as recommended by Cong et al. [44], Cong et al. [10], and Zhang et al.[45].

\[ PAI = \frac{Pen_{after\ aging}}{Pen_{before\ aging}} \] (2)

\[ SPI = SP_{after\ aging} - SP_{before\ aging} \] (3)

Where: \((PAI)\) is penetration aging index, \((SPI)\) is softening point index.

4. Results and discussion

4.1. Penetration

The penetration test gives an indication about the hardness of the bitumen, the results of penetration depth for neat binder (B0) and both w-PF and R-LDPE modified binder are illustrated in Figure (3). Results show that the penetration decrease with the increase in the amounts of modifiers until reached 16 dmm (which represents a reduction of 62.6% compare to B0) at 3.7% MC. Where the penetration decreased by about 24% compare to B0 due to the addition of 3% of R-LDPE, this is due to the swelling phenomena that are resulted from the diffusion of maltenes (oil fraction of binder’s) in the polymeric phase, in addition to the interactions between the polar molecules of asphaltenes and polymer which causes to enhance asphalt polarity properties and forming a polymer network as confirmed by Dehouche et al. [46], and agreed by Yan et al. [47].

The penetration values also showed a noticeable reduction with increasing in w-PF content. Where the ratio of reduction reached 45% at 0.7% w-PF. This behavior can be attributed to that the chemical composition of paper which has a high amount of CaO and SiO₂ that resulting in stiffer asphalt mastic on one hand and to the enhance of rheology by the physical action of fiber on other hand. Therefore, the resistance of the binder to mechanical damages was enhanced [22, 48]. The trend of penetration depth appears in this study is similar to that obtained by Chen et al. [49] when using corn stalk fiber as an asphalt modifier.

The utilization of MC (i.e., R-LDPE with w-PF collectively) to modify asphalt binder shows a significant reduction in penetration values, where it decreased to about 62% when using 3.7% MC (i.e., with the increment of w-PF content). This reduction gives an indication of the improvement in shear resistance of asphalt binder to high temperatures as mentioned by Al-Hadidy et al. [50].
4.2. Softening point (SP)

The verification of asphalt binder consistency was done depending on the results evaluated from softening point test. The results of SP for binder before and after modification are represented in Figure (4). It is clear from the obtained results that the resistance of binder to deformation is enhanced after using the modification materials, which is agreed by a previous study [51]. Where the amount of increment in SP reached 20% after using R-LDPE only in contrast with B0. This increment may be associated with the improvement in asphalt binder structure properties, further to the network properties of R-LDPE polymer as indicated by Eme and Nwaobakata [52].

Results in Figure 4 also indicate that the incorporation of w-PF into asphalt help in increase softening point to the level of 30% at 0.7% w-PF compared with B0. This return to the presence of silica particles in the chemical composition of w-PF that is work on gained asphalt some rigidity. Furthermore, the high surface area and porosity nature of w-PF modifier tends to absorb the lightweight bitumen more. Also, the presence of fiber in the binder structure could form a network that reinforces the binder itself. The results reconcile with that obtained by Mohammed et al. [40] when using cellulose fiber.

The results also displayed that the increase of MC (R-LDPE and w-PF collectively) appear more rising into softening point (i.e., as w-PF dosage increase and fixing R-LDPE dosage). Where the amount of rising reached higher than 60% when using 3.7% MC as shown in Figure 4. This behavior is attributed to the combined effect of the presence of R-LDPE and w-PF into asphalt medium depending on the reasons indicated above for each one.
4.3. Temperature susceptibility

The results of the susceptibility of asphalt binder to temperature were evaluated depending on the index of penetration as demonstrated in Figure 5. Results, in general, indicate that the sensitivity of asphalt to temperature is enhanced after using modifiers. This means that the resistance of the asphalt to rutting and low temperature cracking increase as indicated by Arabani et al. [53]. The usage of R-LDPE resulted in a significant reduction in temperature susceptibility compared to B0. Where the amount of reduction difference 1.305 returns to the increment of the elastic behavior of asphalt binder after comprising R-LDPE due to the formation of the polymer network. A similar finding was indicated by Al-Hadidy et al. [50].

Similarly, as the amount of w-PF increase, the value of PI decreased and the amount of reduction difference reached 1.298, as can be seen in Figure 5. This related to the chemical composition of w-PF that contains SiO₂ particles plus the other effects mentioned previously, which in turn help in increase the binder flexibility then the sensitivity of binder decreased, which is agreed with other findings [22]. The incorporation of the MC into asphalt binder also gave a higher improvement in temperature susceptibility, where PI reached +1.044 at 3.7% MC. This returns to the combined effect of R-LDPE and w-PF which are helpful in extra reduction in the asphalt binder temperature sensitivity.

Figure 4. Softening point of neat and modified asphalt binder, °C

Figure 5. Penetration index of neat and modified asphalt binder, °C
4.4. Ductility

The variation in ductility level of all types of asphalt binders is illustrated in Figure (6). It can be observed that the ductility of the asphalt binder decrease with respect to the increment in the additive’s dosages. The modification of asphalt using 3% R-LDPE lowered the ductility by about 43% compared to B0. This return to the polymer network that increases the binder stiffness then contributes in that reduction. Figure 6 shows as well, that incorporating w-PF resulted in an amount of decrement in ductility level to higher than 50% at 0.7% w-PF. The usage of w-PF gains the asphalt some rigidity due to the presence of SiO$_2$ particles into its chemical composition as confirmed by Chew et al. [22], plus other reasons mentioned in the previous sections. However, the ductility trend appears similar to that observed in the study of Mohammed et al. [40] when using cellulose fiber to improve the properties of asphalt binder. Moreover, the results indicate that the use of MC as a modifier gives more reduction in ductility level reached to 80% at 3.7% MC. This return to the combined effect of these materials will be led to a series of chemical reactions with the chemical components into asphalt molecule then results in more reduction in ductility as shown in Figure 6.

![Figure 6. Ductility of neat and modified asphalt binder, mm](attachment:image)

4.5. Rotational viscosity

The results of the rotational viscosity test for all types of asphalt binders are illustrated in Figures 7, 8 and 9. SHARP recommended that for practical applications to achieve the required workability and pump ability of asphalt, the viscosity should be no more than 3000 centistokes at 135 °C [54]. Generally, the results show that the values of viscosity decreased as test temperature increase. It displayed as well, that the viscosities of asphalt binder with modifiers higher than that without them and all viscosity levels sustain SHARP recommendations (i.e., less than 3000 centistokes). Results present in Figure (7) indicate that the amount of viscosity increased by 1.5 times compared to B0 when using 3% R-LDPE. This is attributed to the same reasons mentioned earlier, where the formation of polymer network help in increase the bond between asphalt molecules plus the change in binder structure from sol to gel-type structure, then resulted in increased asphalt stiffness, as well as, its viscosity. These results are identical to those observed by Punith and Veeraragavan [55].

Furthermore, Figure (8) shows that the incorporation of w-PF into asphalt binder leads to an increased viscosity amount by about 1.7 times in contrast with B0. This attributed to the presence of SiO$_2$ particles as mentioned earlier, as well as, the porosity nature, high surface area, and reinforcement ability of w-PF. All these factors work combined to increase the asphalt binder flexibility and as well
stiffness then help in increasing the level of asphalt binder viscosity to that level. Mohammed et al. [40] in their study show similar results.

The same trend for viscosity is observed when using the MC from R-LDPE and w-PF together. As it can be seen in Figure (9), the level of viscosity increases as MC increases until achieves a rising level equal to 2 times the neat bitumen. This is attributed to the combined effect of R-LDPE polymer and w-PF properties that are mentioned earlier. Then in turn gained asphalt more elastic and rigid which leads to an increase in the viscosity level to this final trend.

**Figure 7.** Rotational viscosity of neat and R-LDPE modified asphalt binder, cSt

**Figure 8.** Rotational viscosity of neat and W-PF modified asphalt binder, cSt
4.6. Aging properties

Figures 10 and 11 show the results of aging indices for asphalt binder before and after modification as a function for penetration and softening point. The results obtained from TFOT simulate the short-term aging that asphalt binder exposed to it during production, mixing and laying. In general, results in Figure (10) display that penetration decreased to 25.8 dmm and SP increased to 55.3 °C when using R-LDPE. The exposure of asphalt binder to temperature in the presence of oxygen leads to a series of chemical reactions into asphalt molecule. The result of this reaction is the increment of the part named asphaltene that is responsible for the hardness of the asphalt binder. Then in turn resulted in a decrease in these fractions at the end, this action works combined with the network formation of R-LDPE polymer and decrease the value of penetration after aging [51].

In the case of w-PF, results indicate that penetration reduced to 20.1 dmm, and the softening point raised to 58.7 °C at 0.7% w-PF. This attributed to the presence of SiO₂ particles, as well as, the porosity nature and high surface area of w-PF, in addition to the action of oxygen and temperature that asphalt binder subjected to it. All combined make the asphalt harder than the results appeared as indicated in Figures 10 and 11.

Moreover, results indicate that the usage of MC shows a similar trend, where penetration and softening point reaches 14.8 dmm and 75.5 °C respectively. The behavior of asphalt binder with MC after aging return to the collective reactions occurred among R-LDPE, w-PF particles and asphalt binder molecules, as well as, the effect of oxygen during the test. Then the influence on the behavior of bitumen after aging appears more than the usage of separate cases between these materials.

Furthermore, the figures show that the factor PAI displays an increment when using modifiers compared to B0, while the factor SPI shows a reverse trend. These factors indices that asphalt binder resistance to aging is enhanced after using the different modification materials with different levels.
5. Conclusion

This study investigated the influence of waste fiber (i.e., w-PF) and recycled polymer (i.e., R-LDPE) on the physical properties of asphalt binder, based on penetration, softening point, viscosity tests and aging index. The following conclusions are offered:

1. The use of w-PF and R-LDPE helps to enhance the physical properties of bitumen significantly in contrast to neat asphalt binder. The collective mix of the modifiers offers better enhancement.
2. Using R-LDPE, w-PF and MC as modifiers decrease penetration up to 24%, 45%, and 62%, respectively.
3. Softening point increases by about 18%, 27%, and 65% for asphalt binder modified with R-LDPE, w-PF and MC respectively.

Figure 10. Aged penetration and PAI of neat and modified asphalt binder

Figure 11. Aged softening point and SPI of neat and modified asphalt binder

Table: Aged Penetration and PAI

| Asphalt binder type | Aged penetration (dmm) | PAI |
|---------------------|------------------------|-----|
| B0                  | 0.76                   | 0.93|
| 3R-LDPE             | 0.90                   | 0.86|
| 0.3W-PF             | 0.75                   | 0.84|
| 0.5W-PF             | 0.85                   | 0.87|
| 0.7W-PF             |                       |     |
| 3.3MC               |                       |     |
| 3.5MC               |                       |     |
| 3.7MC               |                       |     |

Table: Aged Softening Point (℃) and SPI

| Asphalt binder type | Aged SP (℃) | SPI |
|---------------------|-------------|-----|
| B0                  | 48.6        | 3   |
| 3R-LDPE             | 55.3        | 3.3 |
| 0.3W-PF             | 53.1        | 3.1 |
| 0.5W-PF             | 57.2        | 3.2 |
| 0.7W-PF             | 58.7        | 2.7 |
| 3.3MC               | 67.6        | 2.5 |
| 3.5MC               | 75.5        | 2.0 |
| 3.7MC               | 75.5        | 2.0 |
4. The comprising of R-LDPE, w-PF and MC with asphalt binder decreases ductility up to 30%, 56%, and 79%, respectively.
5. The level of viscosity is raised to 1.2, 1.7 and 2 times after the addition of R-LDPE, w-PF and MC, respectively.
6. The susceptibility of asphalt binder to aging is enhanced from 0.76 for neat asphalt to 0.8, 0.86 and 0.93 in terms of penetration index and by 3.3, 2.7 and 2.5 in terms of softening point difference when R-LDPE, w-PF and MC are incorporated, respectively.

According to the results obtained from the improved asphalt, the authors found that these types of additives can improve the performance of asphalt mixtures such as porous and stone mastic asphalt mixture.

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