Optimization the Physical Properties of Waste Denim Fiber Modified Bio-asphalt Binder Using Response Surface Methodology

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Abstract: This research used waste denim fiber (WDF) as a new modifier to mitigate the inconsistency problem of bio-asphalt that incorporated waste cooking oil (WCO). Response surface methodology (RSM) was applied to design and analyze the experimental data. 16 blends of base and bio-asphalt binders were prepared with high shear mixer and evaluated by conducting penetration, ring and ball temperature and temperature susceptibility tests. Optimization of the properties was performed by RSM to come up with the optimum amount of the additives that can be recommended. Results showed that the addition of waste denim fiber reduced the penetration values and increased the ring and ball temperature and the penetration index of base and WDF-modified bio-asphalt binders. These findings reveal that WDF-modified bio-asphalt binders can perform well at the intermediate and high-temperature pavement applications. Numerical optimization results utilizing the developed models exhibited that optimum properties can be achieved with 5% of WCO and 6% of WDF.

1. Introduction
Researchers and asphalt binder industries are always looking for more sustainable alternatives to enhance the performance of the asphalt and minimize its carbon footprint [1-2]. Biotechnology is one of those alternatives which is considered essential for a wide extent of engineering applications [3-4]. Bio-asphalt technologies are also one of the main areas of interest within the field of asphalt pavement engineering for its renewability, cheapness since they are locally sourced, environmentally friendly, and less energy-consuming compared to conventional petroleum products [5]. Many attempts have been conducted to utilize the bio-asphalt binder as modifiers, extenders or replacement for asphalt binder.

Waste cooking oil (WCO) showed the second bio-material was interesting among researchers and succeed to be used as a bio-binder around the world after the swine manure with certain challenges. Malaysia is the fifth ranking of the highest countries in producing and consuming cooking oil around the world [6]. About 160 million gallons annually of WCO in Malaysia, and 6 billion gallons around the world. Therefore this huge toxic waste of WCO needs to be utilized in different applications such as in pavement, otherwise serious environmental pollution will happen [7]. WCO as a bio-waste material
is very cheap compared to the other bio-oils that need pyrolysis and pre-treatment for being used in bio-asphalt. Meanwhile, edible biomaterials should be reserved for the next generations.

Based on the previous research results that applied the WCO as modifier or extender in asphalt binder, there is some achievement was reported in the level of thermal cracking and high-temperature performances [4, 8]. In general, it can be stated that a small replacement amount of 5% of WCO was successfully replaced the conventional asphalt with maintaining similar performance. Therefore, looking for suitable modifiers that can reinforce the WCO before its usage in asphalt binder becomes necessary to improve the possibility of using such bio-waste material for high-temperature applications and to help in solving the environmental issues due to such toxic waste materials. In this regard, WDF is one of the candidate materials to be used to enhance the mechanical characteristics of the bio-asphalt incorporating WCO.

The WDF is mainly produced from the textiles that have been utilized then disposed, the waste of clothes during the production process and the textile goods that have not been sold [9, 10]. The production of cotton was estimated to be more than 20 million tons annually worldwide which is mainly used for producing denim garments. Denim waste fiber contains a high amount of cellulose which up to 95% and 5% of the waxes, nitrogenous matter and pertinacious substances [11-12]. It was claimed that the fibers produced from the denim waste garment showed better mechanical performance compare to the fibers produced from the wood pulp [13]. It was also reported that the WDF has higher specific gravity and molecular weight. In addition to that, it showed significant tensile strength performance compared to the lyocell fibers [13]. The mentioned properties in this finding are considering a higher encouragement to use the waste garment fiber to solve a bio-binder lower molecular weight issue then improve the viscosity and rutting resistance [14-15]. Therefore, the WDF was used and evaluated in this study as a new modifier for enhancing the empirical properties of bio-asphalt binders incorporating WCO.

2. Materials and modification process

2.1. Materials

The penetration grade 60/70 of base asphalt was utilized in this research and provided by PETRONAS refinery, Malacca, Malaysia. The waste cooking oil utilized in this research was obtained from domestic WCO without cost. The domestic WCO was filtered to separate the suspended particles. The WDF was collected from the waste of blue Jeans labeled as 100% cotton and a knife mill was used to reduce the size of WDF with an average of 10 cycles of 15 s.

2.2. Modification process

Base asphalt was kept in the oven at 160°C for one hour to ensure it is fluid enough to pour. Waste cooking oil was used at 0, 5, 10, and 15% by the total weight of blend which was modified by different WDF contents of 0, 2, 4 and 6% by the total weight of the blend. Composite WCO / WDF was heated in a heater at 130°C for a half-hour to allow for WDF to melt in WCO and improve its viscosity before the addition to base bitumen. Then the blends of WCO/WDF were added to the virgin asphalt and the final mixtures were mixed by a high shear mixer at 130°C for one hour and a mixing rate of 3000 rpm to come up with a homogenous bio-modified asphalt binder. The blending parameters of modifiers and base asphalt in this study were obtained by the trial and error method.

3. Experimentation method and analysis

3.1. Penetration test

The penetration test was carried out in accordance with ASTM D5-13 - 2013 standard under a load of 100g for 5 seconds at 25°C to ensure the consistency of unmodified and modified binders. The penetration value was determined as the distance in a tenth of a millimeter.
3.2. Softening point temperature test
The ring and ball temperature test was conducted based on ASTM D36 (2009). The aims of this test to assess the behavior of control and modified bitumen at increased temperatures. The binders were slowly changed from brittle to softer. Ring and ball temperature was taken as the temperature at which the base or modified binder cannot support of 3.5g steel ball at the regular temperature rate (5°C /min).

3.3. Temperature susceptibility
To evaluate the sensitivity of the control and modified bitumen to temperature, the penetration index (PI) parameter was used. Penetration and softening point (ring and ball temperature) tests results were utilized for estimating PI according to Equation (1) as shown below:

\[
PI = \frac{1952 - 500 \times \log(Pen) - 20 \times SP}{50 \times \log(Pen) - SP - 120}
\]

Where pen is the penetration at 25 °C and SP is the ring and ball temperature in °C.

3.4. Design of experiment and statistical analysis
In this study, the user-defined design approach was used by Design Expert 10.0.08 software to design the experiment and perform the analysis. The independent variables in this research were waste cooking oil at four concentration levels of 0, 5, 10 and 15% and waste denim fiber also at four concentration levels of 0, 2, 4 and 6% by the total weight of the blend. The responses were penetration value (dmm), softening point (°C) and penetration index. For each response, 16 samples were prepared according to RSM design. Analysis of variance (ANOVA) was conducted to evaluate the interaction between the independent parameters and the appropriateness of the model. The correlation coefficient (R\(^2\)) was utilized to assess the fitness of the actual responses obtained from the experimental work to the proposed models. Fisher’s test was also employed for ensuring the probability within the confidence level of 95%.

Adequate precision (A.P.) was also utilized to assess noise ratios. The spread of data from its mean value was also evaluated by the standard deviation (S.D.). A coefficient of variance (C.V.) was applied to ensure the reproducibility of the proposed models. Finally, a numerical optimization method was applied to obtain the optimum combination of the mentioned materials to enhance the empirical characteristics of bio-asphalt binders. The numerical optimization criteria are shown in Table 1.

| Parameter          | Unit | Desired objective | Lower limit | Upper limit |
|--------------------|------|-------------------|-------------|-------------|
| WCO                | %    | In range          | 0           | 15          |
| WDF                | %    | In range          | 0           | 6           |
| Penetration        | dmm  | Minimize          | 23          | 364         |
| Softening point    | °C   | Maximize          | 29          | 59          |
| Penetration Index (PI) | -     | In range          | -2          | +2          |

4. Results and discussions
4.1. Penetration test
Penetration can indicate the degree of consistency and stiffness of base and modified binders. Figure 1 illustrates the penetration of composite WDF/WCO modified asphalt binders at 25 °C. It is noted that the addition of WCO leads to a significant increase in the penetration values of bio-asphalt, while the addition of WDF reduced the penetration values of WDF-modified binders and the lowest penetration value was shown at 6% WDF content. It is also observed that the composition of WCO and WDF at various dosages highly decreased penetration values compared to WCO-modified binders. This finding indicates that the use of WDF and WCO/WDF composite could enhance the stiffness and consistency of asphalt binder which is considered the first indicator for the applicability of using such modifiers for
improving the performance of base and bio-asphalt binders, particularly at intermediate-temperature applications.

4.2. Softening point test

The viscosity of asphalt and bio-asphalt binders can be predicted by softening point temperatures. Figure 2 exhibits the softening point temperatures of unmodified and WDF/WCO modified asphalt in °C. In general, it can be observed that there is a significant reduction in the softening temperatures as the content of WCO increases over the various WDF content. On the other hand, it can be noted that there is a remarkable increase in the softening temperatures of WDF/WCO composite binders with an increase of the WDF content up to 4%. This means that the composite WDF/WCO leads to increasing the softening point values of modified binders compared to WCO modified asphalt binder. These results reveal that WDF is a useful modifier to enhance the viscosity of asphalt and bio-asphalt binders. These findings also indicate that the WDF could be a better alternative for enhancing the rutting resistance of modified asphalt binders.

4.3. Temperature susceptibility

Penetration index (PI) is an indicator that is used to explore the temperature sensitivity and flexibility of asphalt binders. The increasing penetration index indicates the higher flexibility of asphalt binders due to the improvement of the gelatine structure of the asphalt matrix. Thus, PI has used to explain the kinds of a colloid binder. For example, when the PI is greater than +2, the binder is considered a gel structure; when PI is lower than -2, the binder is considered as sol structure; others can be considered gel-sol structure. Figure 3 shows the PI values versus WDF contents at various WCO concentrations in modified and bio-modified asphalt binders. It can be observed that PI values of WDF-modified asphalt significantly increases with the addition of WDF compared to the control binder which could be ascribed to the improvement of the binder stiffness. In contrast, WCO has significantly reduced the PI of bio-modified asphalt binders. It can be stated that WDF and composite WDF/WCO significantly improved the PI of modified and bio-modified asphalt binders. These results indicate that composite WDF/WCO modified binders are most likely to perform well at the high-temperature pavement applications.

4.4. Statistical analysis and optimization

After the physical properties’ experiments were conducted in the laboratory, statistical analysis was carried out to have a good understanding of the base and bio-asphalt binders. Regression analysis was used, a fitted quadratic, cubic and quadratic polynomial models were suggested for prediction of
penetration value, softening point, and penetration index respectively. Models were selected based on the suggestion of the software to be significant and to avoid any aliased by the software.

Table 2 shows the statistical analysis summary for responses analyzed in this study. The correlation coefficient ($R^2$) is used to check the degree of correlation and the adequacy of the models. As shown in Table 2, the Penetration value has an $R^2$ of 0.9794, while the softening point and penetration index have $R^2$ values of 0.9886 and 0.7619 respectively. Accepted $R^2$ values obtained for penetration and softening point indicate that there is an adequate relationship among the actual and predicted results. However, for the penetration index, the lower $R^2$ indicates the lower agreement among the actual and predicted values which most probably due to PI is the combination of penetration and softening point. In addition to that, for all models, their adjusted $R^2$ agrees with their predicted $R^2$ because the differences are less than 0.2 [16].

The degree of correlation and adequacy of the models are also checked by evaluating the F and P values as shown in Table 2. The high F values (i.e. 94.98, 57.76 and 12.80) of the models indicate that the model terms are statistically significant for the responses. Also, the lower P values of less than 0.05 for the models indicate that WCO and WDF have significant effects on physical characteristics of control and bio-modified bitumen binders at 95% confidence. Furthermore, adequate precision (AP) is checked to investigate the satisfactoriness of the developed models. For all models, AP values are greater than four which indicates that model adequacy and significant signal to navigate the design space defined by the User-defined design approach.

![Figure 3 Penetration indices of modified bitumen.](image)

| Parameter | Penetration Model | Softening Point Model | PI model |
|-----------|-------------------|-----------------------|---------|
| $R^2$     | 0.9794            | 0.9886                | 0.7619  |
| Adj. $R^2$| 0.9691            | 0.9715                | 0.7024  |
| Pred. $R^2$| 0.9607            | 0.8634                | 0.5477  |
| AP        | 31.533            | 24.826                | 11.702  |
| Std. Dev. | 0.15              | 0.038                 | 0.80    |
| C.V., %   | 3.20              | 1.02                  | 44.22   |
| P-value   | $<0.0001$         | $<0.0001$             | $<0.0001$|
| F-value   | 94.98             | 57.76                 | 12.80   |
The 2D and 3D plots of penetration values, ring and ball temperature and penetration index are presented in Figures 4 and 5 respectively.

As shown in Figures 4a, b, and c, the contour lines are curves, which means there is sufficient interaction among the independent factors [17-18]. From plots in Figure 5, it can be seen that WCO has negative effects on the physical characteristic of asphalt binder because it increases the penetration grade and PI values and reduces softening point. However, adding WDF to the bio-asphalt binder improves the physical properties in terms of penetration grade and temperature susceptibility. Also, it can be seen that softening point response is highly influenced by WDF, and as the WDF percentage increases the ring and ball temperature value increases which reveal the improvement of bio-asphalt binder characteristics at high temperature. Based on this analysis, 6% of WDF showed the lowest penetration value and 4% of WDF showed the highest softening point.

![Figure 4. Contour plots - a) Penetration value b) Softening point, c) Penetration Index.](image-url)
Optimization was conducted based on the criteria presented in Table 1. Based on the higher desirability, it was found that the 5% of WCO and 6% of WDF can be the optimum dosage for improving the physical properties of bio-modified asphalt in this study.

5. Conclusions
This study evaluated the influence of WDF on the physical characteristics of base and bio-asphalt binders that incorporate WCO. Materials were characterized to ensure fulfilling the specifications. Trial and error method applied to design the mixing temperature, rotations per minute and period of mixing that suitable enough for this study. Physical properties tests were carried out to investigate the influences of extender and modifier materials on the characteristics of modified and bio-modified asphalt binders. Design-Expert software applied to design the experiment, develop the models and optimize the physical characteristics of bio-asphalt binder. Results showed that Penetration values decrease and softening point increases with increases WDF content in modified and bio-modified asphalt binders. This result reveals that WDF could enhance the stiffness and consistency of asphalt and bio-asphalt binders which is considered the preliminary indicator for the applicability of such modifiers to improve the bio-asphalt pavement performance. It was also found that the penetration indices enhanced which indicates the lower temperature sensitivity of modified and bio-modified asphalt binders and they are most likely to perform well at the high-temperature pavement applications. From RSM analysis and optimization, 5% of WCO and 6% of WDF can be considered the optimum content for improving the consistency of asphalt binder.
6. References

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