Lawele Granular Asphalt (LGA) is a Buton asphalt product derived from the Lawele area in Central Sulawesi, Indonesia. Although it possesses great potential, LGA utilization has not been fully maximized. One of the challenges is the need for a modifier to extract asphalt from the minerals within LGA. Candlenut oil is a potential modifier that can be used with LGA in Cold Paving Hot Mix Asbuton (CPHMA) due to its similar polarity. Therefore, the aim of this research is to evaluate the performance of Lawele Granular Asphalt (LGA) with candlenut oil as a modifier in Cold Paving Hot Mix Asbuton (CPHMA).

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The Marshall test was conducted to assess the performance of CPHMA with candlenut oil as a modifier. Furthermore, several variations were examined, including the quantity of the modifier, duration of heating and compaction, heating temperature, and storage duration, using seven different mixtures and three storage periods. The optimal composition produced a Marshall value of 687.68 kg, which increased with a longer mixing duration and higher heating temperature.

The results showed that the Marshall value met the standards for CPHMA in Indonesia, as well as for VIM, VMA, VFB, and Flow values. The low flow indicated the density of the CPHMA pavement, while the MQ value showed its ability to withstand deformation of 200.13 kg/1 cm. The behavior of Marshall resistance was supported by Fourier transform infrared (FTIR) spectra, which exhibited similar compound groups between asphalt and the leached results of LGA with candlenut oil, indicating the presence of asphalt (binder). There was a slight decrease in the Marshall value after a 7-day duration, showing an increase after a 21-day storage period. Therefore, candlenut oil served as a viable alternative modifier for LGA

Keywords: Marshall value, Lawele granular asphalt, candlenut oil, asbuton, modifier for Buton asphalt UDC 625 DOI: 10.15587/1729-4061.2023.292080

# OPTIMIZATION OF LAWELE GRANULAR ASPHALT (LGA) PERFORMANCE IN COLD PAVING HOT MIX ASBUTON (CPHMA) WITH CANDLENUT OIL MODIFIER

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Received date 04.09.2023 Accepted date 13.11.2023 Published date 22.12.2023

#### 1. Introduction

Petroleum asphalt is the primary choice for road construction due to its conventional usage and limited availability. However, the need to conserve petroleum asphalt has prompted extensive research on alternative materials, focusing on optimizing local resources. One such material is Lawele Granular Asphalt (LGA) [1], derived from Buton asphalt (asbuton) found on Buton Island, Indonesia. LGA is perceived as a viable option for road construction, offering a sustainable solution by utilizing domestic raw materials.

According to data from 2018 [2], Indonesia typically demanded approximately 1.39 million tons of asphalt annually, leading to the import of 1.04 million tons each year to meet this demand. However, assuming this demand was fulfilled using asbuton, it would require 5.56 million tons/year and an asphalt content of 25 %. From an economic perspective, importing petroleum asphalt would cost approximately US\$436.8 billion/ton (assuming the price of asphalt is \$420/ton [3]), while the use of asbuton leads to a revenue creation of US\$275.5 billion/ton, assuming the price of Buton asphalt is US\$49.56/ton [4]. The cost of applying the wear layer in the field is estimated to be \$83.84/ton for Cold Paving Hot Mix Asbuton (CPHMA) [5], as well as \$109.32/ton and \$94.57/ton for Buton and petroleum asphalt AC-WC, respectively [6]. In terms of CPHMA products, the use of Buton asphalt is more economically beneficial, compared to petroleum asphalt. Regarding the use of AC-WC products, the cost of Buton asphalt is slightly higher than petroleum, which is approximately 0.9 %. These findings suggest that its use tends to provide economic benefits, particularly in the Buton region of Indonesia.

Technologies, 6 (6 (126)), 51-61. doi: https://doi.org/10.15587/1729-4061.2023.292080

How to Cite: Khamelda, L., Djakfar, L., Wisnumurti (2023). Optimization of lawele granular asphalt (LGA) perfor-

mance in cold paving hot mix asbuton (CPHMA) with candlenut oil modifier. Eastern-European Journal of Enterprise

The Food and Agriculture Organization (FAO) stated that different sectors contribute varying percentages of carbon emissions to the environment. For instance, agriculture, industry, transport, building and energy account for 24 %, 21 %, 14 %, 6 %, and 35 % [7]. While within the energy category, coal, natural gas, and oil contribute 43 %, 36 %, and 20 % of carbon emissions, respectively [7]. Asphalt, derived from the final product of distillation, falls under natural gas and oil. Although the specific environmental impact related to the use of Buton asphalt has not been extensively studied, the overall impact can be evaluated from raw material processing to its application in the field. Compared to the lengthy processes involved in oil distillation, the mining process for Buton asphalt is less complex. Once mined, Buton asphalt can be directly used in certain pavement applications. The technique used to process it into raw material for pavement types that utilize full or semi-extracted asbuton involves a shorter process than the production of petroleum. Buton asphalt requires less energy during processing compared to petroleum, thereby resulting in reduced carbon emissions from the industrial sector [8].

Therefore, studies that are devoted to the development of buton asphalt technology are scientifically relevant to the needs of transportation facilities today and in the future, because the need for road construction will grow as the number of people and settlements develop. So it takes ongoing research to be realized in the field.

#### 2. Literature review and problem statement

Currently, LGA is mostly used as an additive or filler to reduce the reliance on petroleum asphalt. However, a significant obstacle in effectively utilizing Buton asphalt as a binder in road pavement mixes is the difficulty in extracting the bitumen stored within the mineral cavities [9]. In order to enable Buton asphalt to act as a permanent binder in road pavements, similar to petroleum asphalt, a modifier is required to dissolve the asbuton bitumen and modify its characteristics [10, 11]. One alternative modifier that has been explored is candlenut oil, which can potentially enhance the properties of Buton asphalt [12].

This is because the bitumen on Buton asphalt is stored in minerals, so a material is required to dissolve bitumen. The solubility of vegetable oils is determined by the polarity properties of fatty acids [13]. Fatty acids are hydrocarbon chains with added carbon dioxide ( $CO_2$ ) at one end. Hydrocarbons located on fatty acid compounds are the compounds with the largest percentage in vegetable oils so hydrocarbons are assumed to play a major role in the dissolution of bitumen.

The LLE [12] test showed a high percentage of bitumen solubility, but the Marshall test stability is still below the CPHMA standard. This suggests that the maximum soluble bitumen may not be caused by the presence of certain substances in the oil or the need to add treatment variations to its application. A modification is needed in the application of candlenut oil to optimize the amount of solubility of asbuton bitumen so that it can produce maximum CPHMA performance. Thus, even though the potential of candlenut oil has been demonstrated in early research [12], further research is needed to maximize CPHMA performance.

This research is not only to demonstrate the capabilities of LGA as a road pavement material but also to investigate the further potential of candlenut oil as an alternative modifier for Buton asphalt. By using candlenut oil as a modifier, the technological advancements of Buton asphalt tend to be improved, while increased utilization potentially benefits farmers, particularly when the oil is produced on an industrial scale.

Several studies have been conducted to obtain a suitable modifier for LGA. The paper [14, 15] showed the advantages of using naphthalene that it produces less porous damage, has higher solubility performance, more efficient and uni-

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form dilution, fewer residues trapped in the pores, minimal asphalt sedimentation, but its weaknesses are that it generates less asphalt, shows a slower rate of deposition, higher surface temperatures. Naphthalene is a polycyclic aromatic hydrocarbon that is widely used in a variety of applications. However, there are potential dangers associated with its use. Critical reviews of naphthalene sources and exposures highlight recent classifications that may be carcinogenic to humans. This review emphasizes the need to understand the source and exposure to naphthalene, given its widespread presence and potential health risks [16]. Moreover, [17] discusses the sources, concentrations, and risks of naphthalene in indoor and outdoor air. The study highlights the presence of naphthalene as a volatile organic compound (VOC) in a variety of environments and potential health risks, including its association with neoplasms and environmental exposure. Overall, this reference highlights the potential hazards associated with the use of naphthalene. These dangers include classification as a possible human carcinogen, risk of absorption and exposure, and potential adverse effects on birth outcomes. It is important to be aware of these dangers and take appropriate precautions when handling and using products containing naphthalene. The paper [18] indicates the advantages of using trichloroethylene, which can help to soften asphalt binders significantly, but its disadvantage is toxicity. Trichloroethylene (TCE) is a solvent widely used in industry. Despite its usefulness, there are some potential concerns and limitations associated with its use. Another concern associated with TCE is the environmental impact. The research [19] states that TCE is a chlorinated solvent that can contribute to air pollution and environmental contamination. The study emphasizes the need for an environmentally friendly and efficient alternative to TCE due to its potential negative impact on the environment. In addition, TCE has been classified as a volatile organic compound (VOC) and is subject to regulations and restrictions due to its potential negative effects on human health and the environment. The research [20] addresses the determination and monitoring of TCE in ambient air, emphasizing the need for accurate analysis and quality control to evaluate TCE levels and ensure compliance with regulatory standards. The paper [17] shows the advantages of trichloromethane, which is widely used, easy to evaporate, reusable, however its weaknesses are harm to the user and the environment, effects on asphalt binders. Trichloromethane, commonly known as chlorophore, emphasizes the potential risks associated with trichloromethanes and the importance of effective degradation methods to reduce their harmful effects. Heckel et al. highlighted the potential environmental impact of trichloromethane and its transformation products. [21-24] identified trichloromethane as a groundwater pollutant in the Junggar Basin in Xinjiang, China, stressing the need for risk assessment and management strategies to protect water resources from contamination. The paper [18] shows the benefits of using carbon tetrachloride, which can prevent the overlap of proton signals because of solvents so that it can help use the solvent mixture, can make the surface voltage of the sample so high that it facilitates the process of sample interaction, not easy to combustion, but its degradation is toxic to organisms, at high concentrations can produce unhygienic phosphene gases. Carbon tetrachloride is a chemical compound that has been extensively studied for potential hazards and risks. These dangers include kidney damage [25], lungs [26], oxidative stress [27] and liver injury [28, 29].

Generally, the papers [14–18] present the results of research on chemical-based modifiers for LGA shown through chemical testing. But there were unresolved issues related to applying the modifier to the pavement sample with mechanical tests. The reason for this may be the toxicity of the chemicals. The price is relatively high, there is no advanced research, etc. A way to overcome these difficulties can be using environmentally friendly materials or mechanical testing application, etc. This approach was used in [30], however, the materials used are not commonly available on the market, and there are no chemical studies of the material's ability as an LGA modifier. All this suggests that it is advisable to conduct a study of asbuton modifier on sustainable modifier material.

#### 3. The aim and objectives of the study

The aim of this study is to optimize the performance of CPHMA with candlenut oil modifier.

To achieve this aim, the following objectives are accomplished:

 to formulate the composition of the CPHMA mixture that will provide optimal resistance in receiving the load tested with the Marshall test;

– to detect the presence of a cluster of LGA bitumen compounds on the extraction material with the FTIR test so that to prove the candlenut oil ability to dissolve the LGA bitumen.

#### 4. Materials and methods

#### 4. 1. Object and hypothesis of the study

The research object is the composition of the CPHMA mixture using a candlenut oil modifier that will produce an optimal CPHMA performance, the performance will be tested chemically and mechanically.

The main research hypotheses are:

 – if the LGA Liquid compound cluster is the same as the oil asphalt, this proves that candlenut oil is capable of dissolving bitumen from LGA;

– if the LGA bitumen can be dissolved by candlenut oil, then the bitumen will bind aggregates that form a hardening that is resistant to pressure loads.

Assumptions adopted in the work are:

 – candlenut oil can dissolve LGA due to the similar polarity with asphalt;

- mechanical tests can be performed if a pavement is formed, which is a bond between bitumen and aggregate.

Simplification adopted in the work is the acquisition of the modifier. The modifiers used are easy to find in the market so they do not extend the production chain.

The modification is studied through the composition of the CPHMA mixture because it requires the right composition to produce maximum performance. The composition includes the quantity of the modifier, the temperature and duration of heating as well as the compression temperature. This is in line with the Gertenbach study (2002), which states that solubility is influenced by the type of solute, temperature, solute quantity and solute particle size [31].

#### 4.2. Materials

#### 4.2.1. Marshall test

The materials used were components of the CPHMA mixture, as shown in Fig. 1. The mixture consisted of

761.1 grams of aggregates that adhered to the CPHMA requirements in terms of gradation [32]. Buton asphalt weighing 238.9 grams contains 60 grams of bitumen [12, 33] and 47.8 grams of candlenut oil [12].





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Fig. 1. Materials of the Marshall test: a - aggregate; b - candlenut oil; c - Lawele granular asphalt

The aggregate was obtained from a stone breaker manufacturer in Malang, Indonesia, who brought the stone from Lumajang, Indonesia, according to gradation based on CPHMA specifications [34]. While LGA from Lawele, Indonesia is obtained from the CPHMA manufacturer located in Pasuruan, Indonesia.

#### 4.2.2. FTIR test

The materials used in the FTIR test were Pen 60/70 Asphalt, LGA Liquid and Solid, LGA 50/30 and bitumen LGA (extracted from LGA using a TCE solvent). Fig. 2 shows LGA Liquid and Solid, resulting from leaching LGA with candlenut oil.



Fig. 2. Materials of the Fourier transform infrared test: a - Lawele granular asphalt liquid; b - Lawele granular asphalt solid

The material tested with FTIR can be either solid or liquid. LGA Solid and LGA are solid materials while others are liquid. Bitumen LGA is obtained from extraction against LGA using TCE as a modifier. An option against TCE because it is a common modifier used in dissolving asphalt. 0

0

0

0

0

0

0

#### 4.3. Methods

#### 4.3.1. Procedure

The testing procedure follows the flow chart shown in Fig. 3. The tests are conducted in two paths, namely the Marshall and FTIR tests. In this study, both paths are performed simultaneously, if done gradually it doesn't matter.

As shown in Fig. 3, the mechanical tests are carried out with the Marshall tests. The materials used are LGA, candlenut oil and aggregate. All three are mixed and compressed with different treatments. This will later show the composition that will give the optimum Marshall parameters. As for chemical testing, it is done with FTIR testing. The materials used are LGA Solid and LGA Liquid. Both are the result of leaching asbuton using candlenut oil. The FTIR test also tested Asphalt Pen 60/70 and LGA bitumen to determine the compound clusters contained for later comparison with leaching material. So that we can identify a group of compounds that identify the presence of bitumen.



Fig. 3. Testing procedure

#### 4.3.2. Marshall test

The Marshall test results are an analogy of the hardening conditions in the field so the test sample needs to meet the Marshall standard. The Marshall test, based on SNI 06-2489-1991 [35], was conducted to obtain parameters that serve as the reference standard for the strength assessment of CPHMA pavement. The test parameters used include Marshall Stability, Flow, Marshall Quotient (MQ), Voids in Mineral Aggregate (VMA), Voids Filled with Bitumen (VFB) and Voids in Mixture (VIM).

In order to comply with the CPHMA requirements in Indonesia, the Marshall parameters need to meet specific thresholds, including Marshall stability>500 kg, flow>3 cm, VMA>16 %, VFB>60 %, 4<VIM<10. Mixture composition variations are shown in Table 1.

The determination of temperature variation is based on the maximum permissible limit for heating the CPHMA mixture, so is the case with the variation of the compression temperature. But the temperature that exceeds the specified 100 °C is tested based on previous research [7] to determine the effect of the increase in the compression temperature on the CPHMA performance. Whereas the variations in the heating duration are based on time efficiency considerations. Variations in the quantity of candlenut oil have been tested in early research [8], and it was found that the use of 10 % modifier cannot dissolve asbuton bitumen optimally while 40 % will cause saturation on the mixture causing the sample to become too moist. Test variations are shown in Fig. 4.

#### **Composition Variations of CPHMA Mixture**

Code	Heat	ing	Compaction	Storage	Candlenut
	Temperature (°C)	Duration (min)	Temperature (°C)	(days)	Oil (%)
OP#1	170	40	100	0	20
OP#2	170	60	100	0	20
OP#3	200	40	100	0	20
OP#4	170	40	100	0	30
OP#5	170	75	50	0	20
OP#6	170	90	100	0	20
OP#7	170	75	25	0	20
V~~#4	170	75	50	7	20
var#1	170	75	50	21	20
V~~#9	170	75	25	7	20
var#2	170	75	25	21	20
Var#3	170	75	25	7	20
	170	75	25	21	20



#### Fig. 4. Variations of Cold Paving Hot Mix Asbuton Mixture

Fig. 4 shows the Marshall test process. At the start, the test was performed against the variation of the mixture OP#1 to OP#7, then the variation showing the highest stability value will continue its test against the duration variable. In Var#1 and Var#3, heating is accomplished simultaneously with mixing and then stored, while Var#2 is stored after mixing at room temperature, with heating carried out before compaction.

#### 4.3.3. Fourier transform infrared (FTIR) test

The FTIR test was conducted to ensure the performance of candlenut oil as an LGA modifier. The objective of the FTIR test was to investigate the presence of asphalt compound groups in the liquid obtained through the leaching process of LGA using candlenut oil. The leaching or centrifugation process is an extraction method aimed at separating liquids from solids by adding a solvent [36]. Based on previous research [12], it is a recommended cold extraction technique for modifiers with high boiling points.

In this research, the leached samples were analyzed using the Perkin-Elmer FTIR spectrometer Frontier instrument (PerkinElmer, Inc., Waltham, MA, USA) to obtain their FTIR spectra. Spectrum scanning was recorded at a resolution of 4 cm<sup>-1</sup>, covering a wavenumber range of 4,000 to 400 cm<sup>-1</sup>. Micro TF fiber samples were mixed with potassium bromide and pressed into pellet-shaped molds for observation.

#### 5. Results of optimizing the performance of CPHMA with candlenut oil modifier

5.1. Performance testing of the CPHMA mixture with the Marshall test

5.1.1. Stability

The stability value is shown in Fig. 5.



Fig. 5. Marshall Stability: a - without storage; b - with storage

The stability value as shown in Fig. 5 can be explained as follows:

a) without storage.

Based on the Marshall test results, it can be observed that temperature greatly affects the Marshall strength. Higher heating durations and compaction temperatures result in increased Marshall values. This indicates the significant influence of temperature on the quantity of asphalt released from LGA minerals. Heating the LGA facilitates the softening of minerals with candlenut oil. This behavior is supported by the higher Marshall values obtained from samples compacted at temperatures of >25 °C compared to those at 25 °C. Similarly, longer heating durations lead to higher Marshall values. While heating temperature and modifier quantity also impact the Marshall value, this research reported that the optimal composition is a heating temperature and modifier quantity of 170 °C and 20 %, respectively. Samples with a heating temperature and modifier quantity of 200 °C and 30 % yield lower Marshall values compared to the optimal conditions;

b) with storage.

The Marshall value is the highest in OP#6, corresponding to the longest heating duration. However, the difference in Marshall values between OP#6 and OP#5 is not significantly notable. Based on the efficiency of heating duration, the optimal mixture tested for storage variations is OP#5. The OP#5 mixture was further treated with variables of storage duration of 7 and 21 days. While OP#7 also continued testing with the storage variable because it varied OP#5 with the compression temperature variable of 25 °C. The Marshall value decreases as the storage duration increases because prolonged storage allows for mineral and trapped asphalt hardening. An anomaly was observed in sample Var#2 during the 21-day period, where the Marshall value slightly increased after a decrease during the 7-day period. This indicates that the heating time, whether before or after storage, does not significantly affect the Marshall value.

## **5. 1. 2. Flow** The flow value is shown in Fig. 6.



Fig. 6. Flow: *a* – without storage; *b* – with storage

The flow value as shown in Fig. 6 can be explained as follows:

a) without storage.

Among all the variations, OP#4 showed the lowest Marshall value and the largest decrease, leading to the highest flow. This can be attributed to the high VIM (Voids in Mineral) value. When the flow value is low despite a relatively high VIM value and the same VFB (Voids Filled with Bitumen) as other samples, it indicates the formation of stronger bonds between the asphalt and aggregates in OP#4. This is expected to be supported by a high MQ (Marshall Quotient) value;

b) with storage.

In general, there is an inverse relationship between flow and the Marshall value, except for day 0, where Var#1, despite having the highest Marshall value, also exhibits the highest flow. On the other hand, Var#2 and Var#3 show similar flow values. Analyzing the three variations at 7 and 21 days of storage, it was observed that the availability of VMA and the filling of VIM differ, while the VFB values remain relatively consistent. The larger the VMA, the greater the VIM values. Assuming this behavior of voids is associated with flow, then at 7 days of storage, Var#3 tends to exhibit denser characteristics than the other variations. This is supported by the low VIM value and flow, suggesting no voids are available for deformation.

#### 5.1.3. Marshall quotient

The MQ value is shown in Fig. 7.

The MQ value as shown in Fig. 7 can be explained as follows:

a) without storage.

Sample OP#5 exhibits the highest MQ value, indicating its superior strength in resisting deformation. It requires a load of 200.13 kg to cause a 1 mm deformation in the pavement. In contrast, sample OP#4 shows the lowest strength, succumbing to deformation with a load as low as 51.46 kg. The MQ value further confirms that OP#5 is the optimal composition variation. With its relatively high Marshall value and exceptional resistance to deformation, it stands out as the most favorable choice because asides from having a relatively high Marshall value and exceptional resistance to deformation, it stands out as the most favorable choice;

b) with storage.

The MQ values for Var#1 exhibit a relatively stable trend, indicating a consistent ability to withstand load even as the Marshall and flow values decrease. There is an overall decline in MQ at 7 and 21 days when considering the storage duration. However, the decrease at 21 days is less pronounced than at 7 days. There is a potential increase in MQ at 7 days of storage for Var#1 and at 21 days of storage for Var#2. The average decrease in MQ is 3.31 % and 1.56 %/day for a 7-day and 21-day storage duration, respectively.



Fig. 7. Marshall Quotient: a – without storage; b – with storage

#### 5.1.4. Voids in mineral aggregate

The VMA value is shown in Fig. 8.

The VMA value as shown in Fig. 8 can be explained as follows:

a) without storage.

The 7<sup>th</sup> sample, known as OP#4, stands out by fulfilling the CPHMA standard with the highest VMA value among all variations. Interestingly, this achievement is not due to differences in aggregate gradation, as all variations used the same gradation. Instead, the lack of density in sample OP#4 significantly influences the VMA value. This can be inferred from the behavior of flow and VIM, both of which show the highest values in this sample. A high VIM value indicates a relatively lower density than the other samples, leading to noticeable deformation. The insufficient density could be attributed to the excessive use of the modifier during compaction, where it fills the voids and increases the moisture content of the sample. Additionally, after a 24-hour period of undisturbed settling, the modifier adheres to the aggregate, creating air voids;

b) with storage.

Var#3 shows the highest VMA value after 7 and 21 days of storage. This outcome can be linked to the timing of heating the mixture, as Var#3 undergoes heating just before compaction. Interestingly, the positions of Var#2 and Var#1 are swapped at 21 days of storage, and the difference can be attributed to the compaction temperature. These observations suggest a clear correlation between the heating time and compaction temperature with respect to the VMA values obtained.



Fig. 8. Voids in Mineral aggregate: a - without storage; b - with storage

### 5. 1. 5. Voids filled with bitumen

The VFB value is shown in Fig. 9.



Fig. 9. Voids Filled with Bitumen: a - without storage; b - with storage

The VFB value as shown in Fig. 9 can be explained as follows:

a) without storage.

The VFB value represents the degree to which voids in the mixture are filled with asphalt. Surprisingly, the VFB values of the 7 samples are relatively similar, despite differences in their VMA values. This indicates that the asphalt released by LGA has been optimized. Although air voids are still available, they are not filled with asphalt. It is important to note that assuming the asphalt content exceeds the capacity of VMA, bleeding may occur because there are no more voids left to accommodate the additional asphalt;

b) with storage.

Samples that underwent storage displayed a similar range of VFB values compared to those that were not stored. This implies no noticeable reduction of asphalt content in the mixture during the storage process. One potential reason for this is the re-absorption of asphalt by the LGA mineral during storage, thereby preventing a significant decrease in the overall content of this element.

#### 5.2.6. Voids in mixture

The VIM value is shown in Fig. 10.

The VIM value as shown in Fig. 10 can be explained as follows:

a) without storage.

No anomalies were observed in the VIM values, as these generally correlate with the VMA. Interestingly, the samples with the lowest VMA quantity also have minimum VIM. This observation is consistent with the similar VFB values obtained across the samples;

b) with storage.

The behavior of VIM values in stored samples is comparable to those not stored.



Fig. 10. Voids in Mixture: *a* – without storage; *b* – with storage

5. 2. Detection of the presence of bitumen compound clusters on extraction material of Lawele granular asphalt with Fourier transform infrared testing

The results of the FTIR test are shown in Table 4 and Fig. 5.

Table 4

Travendiliber											
Wave (cm <sup>-1</sup> )						Eupetional Croups	Suspected Compound				
Asphalt Pen 60/70	LGA Liquid	LGA Solid	Candlenut Oil	LGA	Bitumen LGA-T	Functional Groups	Cluster				
0	0	0	0	3625.43	~3,600	O-H	Hydroxyl				
~3,500-3,900	~3,500-3,900	0	~3,500-3,900	0	~2,000-2,300	С-Н	Aromatic				
0	0	3,405.40	0	3,401.51	0	N–H	Amine				
0	3,009.31	3,006.45	3,009.31	0	2,953.68	C–H (sp <sup>3</sup> )	Alkane				
2,922.31	2,923.73	2925.16	2,923.73	2,925.16	2,922.31	C–H (sp <sup>3</sup> )					
2,852.42	2,855.28	2,853.85	2,853.85	2,855.28	2,855.28	C–H (sp <sup>3</sup> )					
0	0	2,512.98	0	2,512.98	0	S-H	Thiol				
~2,000-2,350	0	0	~2,000-2,300	0	0	С-Н	Aromatic				
1,744.26	1,741.40	1,744.26	1,742.83	1,798.45	1,727.14	C=O	Aldehyde				
~1,500-1,650	~1,500-1,652	1,575.96	~1,500-1,652.98	0	~1,500-1,600	C=C	Aromatic				
0	0	1,426.21	0	1,424.79	0	O-H	Hydroxyl				
1456.16	1,457.59	0	1,460.44	0	1,457.59	С-Н	$CH_2$				
1373.44	1,374.87	0	1,373.44	0	1,377.73	С-Н	$CH_3$				
0	0	0	0	0	1,269.33	C–O	Alkyl Aryl Ether				
1160.94	1,160.94	0	1,162.36	0	0	C–O	Ester				
1029.72	1,099.61	1,035.43	1,098.18	1034.00	1,071.08	S=O	Sulfoxide				
0	0	874.21	0	874.27	0	C=C	Alkene				
724.52	718.81	711.68	720.24	710.25	741.63	С-Н	_				

Wavenumber



Fig. 11. Fourier transform infrared transmission

The compound groups found in LGA Liquid exhibit a close resemblance, almost 100 %, similar to Pen Asphalt and LGA Bitumen. This similarity indicates the presence of bitumen in LGA Liquid, suggesting that it has been dissolved by candlenut oil during the leaching process. The compound groups in LGA Liquid also bear a striking similarity, almost 100 %, to candlenut oil. This similarity suggested that the solvent was mixed with bitumen at the end of the leaching process. Regarding LGA Solid, its compound groups are identical to those found in LGA, indicating the presence of predominantly mineral compound groups. Since it consists of 75 % mineral content, this finding aligns with the mineral composition of LGA. LGA Solid exhibits slight variations in compound groups compared to Pen Asphalt and LGA Liquid. The presence of Amine (N-H), Thiol (S-H), and Alkene (C=C) compound groups originated from LGA minerals. The absence of Aromatic (C-H), C-H<sub>3</sub> (-CH<sub>3</sub>), and Ester (C-O) indicates a lack of bitumen.

## 6. Discussion of candlenut oil performance on Lawele granular asphalt

Fig. 5 shows that the Marshall stability value of the CPHMA mixture experiences an increasing trend with the addition of asphalt content until the optimum asphalt content (OAC) is reached. However, exceeding the OAC leads to a decrease in the Marshall value [37]. It is crucial to ensure that the asphalt content derived from LGA minerals aligns with the OAC. To investigate the quantity of LGA asphalt in the CPHMA mixture, variations in testing are conducted on variables that are assumed to have an impact.

Fig. 6 shows that the flow value is influenced by the voids present in the mixture, particularly the quantities of VIM and VFB, and not just the availability of VMA. When a sample has a relatively high VIM value, it indicates many voids in the mixture filled with air. As the VIM value increases, the sample becomes less capable of withstanding pressure and more susceptible to deformation [38].

Fig. 7 shows the MQ value, it is an indicator of the actual strength of the sample in terms of its ability to resist load. This represents the load value that can be sustained for each 1 mm of deformation. It is important to note that the strength of the sample cannot be accurately determined by the Marshall value alone. In theory, the Marshall value is directly proportional to the flow. Logically, the longer the sample can withstand the load until failure, the higher the Marshall and flow values. It is common to observe cases where the Marshall value is high while the flow is low or vice versa. Relying solely on the Marshall value may not accurately reflect the strength of the pavement in accepting the load. For a comprehensive understanding of the sample strength, it is necessary to consider multiple parameters, including the MQ value.

Fig. 8 shows that the VMA value exhibits a distinct behavior. Initially, it decreases until it reaches a minimum value, after which it starts increasing with the addition of asphalt content [37, 38]. In circumstances where the VMA value is below the standard, the sample becomes overly dense due to the voids being filled with asphalt, displacing the air and reducing the VIM. This behavior shows the impact of sample density on the VMA value. Furthermore, the density is also influenced by aggregate gradation. Uniformly and close-graded aggregates tend to yield the largest and smallest VMA, respectively.

Fig. 9 shows the VFB value that refers to the percentage of voids between aggregate particles filled with asphalt, excluding the absorbed ones [39]. This parameter is significant in assessing pavement durability, as a higher VFB value indicates improved pavement impermeability [38, 40]. As the asphalt content increases, the VFB value also increases, indicating a greater filling of voids with asphalt [37].

Fig. 10 shows that the VIM value decreases as the asphalt content increases since it fills more voids, reducing the presence of air [37, 41]. A smaller VIM value indicates a denser sample prone to plastic deformation and compaction [38]. However, a lower VIM value also increases the potential for bleeding in the pavement due to a lack of air voids that can accommodate excess asphalt [38, 39]. A higher VIM value can lead to oxidation or ageing of the asphalt due to reduced impermeability caused by air infiltration and sunlight exposure [39, 40]. As the asphalt content in the mixture rises, the VFB value also increases, resulting in a smaller VIM value [37]. This is also influenced by the quantity of VMA.

Table 4 and Fig. 11 show that the potential of candlenut oil as an asbuton modifier has been demonstrated through the LLE test [12] and confirmed by the FTIR test. The results of the FTIR test have shown the presence of bitumen from LLE tests so it can be said that candlenut oil can dissolve bitumen on LGA. This is in accordance with the principle of "like dissolves like" where materials with the same properties will be able to solve each other [42]. The solubility is due to the similarity between vegetable oil and asphalt as non-polar compounds [42–44]. In addition, both are organic materials in which organic material can be soluble in organic solvents [28].

This study proved the potential of materials that are capable of becoming a modifier of LGA, in particular CPHMA, which is vegetable oil. The use of vegetable oil as a sustainable material in LGA products has been carried out in the research [30]. But in such research, the materials used are not easily found on the market, or require special processes to produce them, which will prolong the production process. As for other studies [14–18] that use chemical-based modifiers, of course, these are not sustainable materials. While they can be produced sustainably, toxic factors are one thing to consider.

The limitation of this study is the lack of references that can be used as a reference in research. This is because LGA-related research is still on the scale of basic research so not much is known. The other limitation is the diversity of research methods that can be applied to CPHMA with a vegetable oil-based modifier. Vegetable oils need high temperatures to boil, so they need equipment to facilitate it.

The weakness of this study is that the quantity of bitumen used in the CPHMA sample has not been proved. Although previous studies [12] have shown the potential of candlenut oil in dissolving the LGA bitumen through chemical testing, in fact it is not yet known exactly how much bitumen forms the CPHMA. This test can actually be done by extraction on the CPHMA samples, but if conventional methods such as Reflux or Soxhlet are used, high temperatures will be required to make the candlenut oil boil and evaporate causing the equipment to be susceptible to rupture. This has been tested on simple patent research [45]. Perhaps, a tool Distillation Of Cutback Asphalts can be used for extraction, which can facilitate the use of high temperatures.

Although the mechanical test results of this study have not shown optimal results, this study has proved the potential of candlenut oil as a modifier of CPHMA, both chemically and mechanically. Ideally, the modifier does not only function as a bitumen solvent but also as a material capable of improving the quality of CPHMA. Based on the results of the mechanical tests in this study, it was found that the longer the storage time, the lower the quality. This is not good. At least even though there is a decrease in the quality but it should not be too significant, because one of the advantages of CPHMA is that it can be stored for up to 6 months in ready-pave condition. Advanced research is needed to be able to find a solution in the use of candlenut oil as a CPHMA modifier.

#### 7. Conclusions

1. The Marshall test on the CPHMA mix shows the optimal stability value obtained from the variation of the mixture OP#5, which is Var#1 with 687.68 kg in 0 storage days. Var#1 gets a compression treatment at the highest temperature compared to Var#2 and Var#3, which is 50 °C. This indicates that the compression temperature affects the resistance of the adhesion in holding the load shown through the stability value. As for other Marshall parameters, there is no difference with Marshall's behavior in general.

2. The FTIR test on Liquid LGA showed the presence of a bitumen compound cluster as shown on 60/70 asphalt and LGA-T bitumen indicated by the presence of asphaltene, aromatic, saturates and resins. Asphaltene is indicated with an absorption of  $1,029 \text{ cm}^{-1}$  (S=O) and 730 cm<sup>-1</sup> (-(CH2)n). Whereas  $3,400 \text{ cm}^{-1}$  (=C-H) indicates an aromatic ring. Resin is indicated at  $690-900 \text{ cm}^{-1}$ , which is the presence of ether (C-O) and alkyl amine (C-N). Whereas on Solid LGA, there is mineral presence as shown in LGA. This proves that candlenut oil is capable of solving against LGA bitumen.

#### **Conflict of interest**

The authors declare that they have no conflict of interest in relation to this research, whether financial, personal, authorship or otherwise, that could affect the research and its results presented in this paper.

#### Financing

The authors would like to thank Brawijaya University for financial support. Grant Number 593.89/UN10.C10/ PN/2020.

#### Data availability

The manuscript has no associated data.

#### Acknowledgments

The authors are grateful to:

- 1. Researcher Team (D. J. Djoko H. Santjojo, Ahmadiansyah, R. Abednego Rumbay, C. Chandra Septyan);
  - 2. PT. KAN for providing LGA.

#### Use of artificial intelligence

The authors confirm that there are no artificial intelligence technologies being used when creating the current work.

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