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## Ductility improvement of high density polyethylene (HDPE)-modified bitumen through adding modified lignin

To cite this article: E A Septimorien *et al* 2019 *IOP Conf. Ser.: Mater. Sci. Eng.* **541** 012035

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# Ductility improvement of high density polyethylene (HDPE)-modified bitumen through adding modified lignin

E A Septimorien<sup>1</sup>, L M D Friskila<sup>1</sup>, B Natasya<sup>1</sup> and M Chalid<sup>1</sup>

<sup>1</sup>Department of Metallurgical and Materials Engineering, Universitas Indonesia, Depok 16424, Indonesia

E-mail : chalid@metal.ui.ac.id

**Abstract.** High Density Polyethylene (HDPE) is added to bitumen to improve the mechanical properties as Polymer Modified Bitumen (PMB), but both of them have different chemical properties. To achieve best mechanical properties of the mixture coupling agent is needed such as lignin that has bipolar characteristic. In the previous research, unmodified lignin tendency of polarity is bigger thus modified with polyurethane PEG to increase its non-polar parts. In order to confirm the potential of modified lignin as a coupling agent for HDPE-Modified Bitumen. Several stages of this experiment consists of sample preparation, characterization of raw materials, mixing, and characterization of the PE-Modified Bitumen. This experiment used hot melt mixing to mix modified lignin, HDPE, and bitumen. The result of this experimental was analysed by using Ductility test, FTIR and FESEM. The addition of modified lignin to HDPE-Modified Bitumen increase distribution and dispersion of HDPE particulates inside bitumen matrix, increasing PMB HDPE mechanical properties. Furthermore, FESEM characterization indicated that the addition of modified lignin gave better mixing of PE-Bitumen. FTIR showed an increase of intensities at non-polar peaks, and down at polar peaks compared with unmodified lignin. Ductility test showed that modified lignin has better compatibility as coupling agent than unmodified lignin. From this experiment, modified lignin can improve mixing between HDPE and Bitumen better than modified lignin. So, we can use modified lignin as the new coupling agent.

## 1. Introduction

The applications of bitumen as matrix material have a weakness such as plastic deformation when receiving pressure such as a vehicle. For countering that weakness, reinforcement should be added as filler. In order to obtain bitumen with better quality, an increasing number of research began to focus on bitumen modification. Bitumen modification commonly used polymer materials. Polymer for bitumen modification include polyethylene waste (WPE), polypropylene, and tire crumb rubber [1-5]. From the research result, it shows a positive effect, which solves the waste problem in the beginning of this part. But when it is viewed chemically, bitumen has a hydrophilic characteristic that is polar and polymer is the opposite, hydrophobic and is non-polar, which make the two the exact opposite making them hard to interact with one another. What needed for the two of them to become one is the presence of coupling agent that acts as a bridge between these two.



## 2. Experimental

### 2.1. Materials

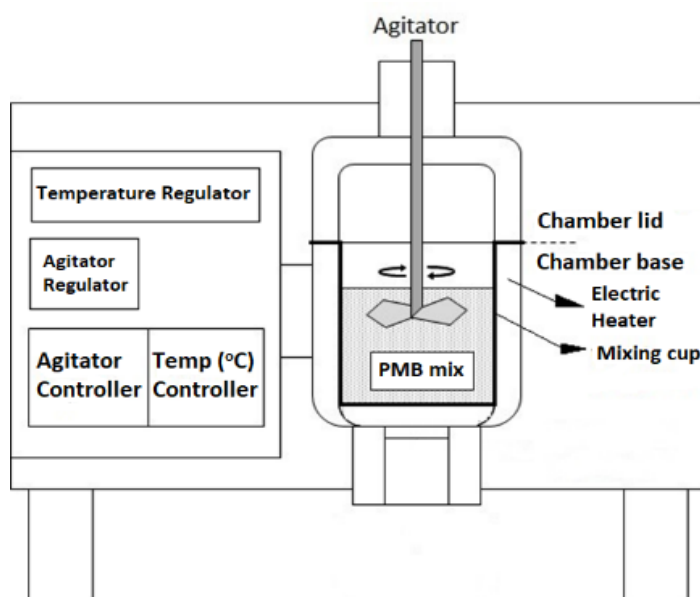
The materials used in this research are HDPE Plastic bags from commercial market. Bitumen come from Pertamina with pen 60/70 grade. While the modified lignin made in accordance to our laboratory<sup>[6]</sup>. With the materials to produce modified tignin are lignin from Technical Chemicals Indonesiaku, 4,4-methylenebis (cyclohexyl isocyanate) (HMDI), polyethylene glycol (PEG) with MW 6000 g/mol, and dibutyltin dilaurate (DD) as catalyst purchased from Sigma Aldrich.

### 2.2. Preparation of HDPE-modified bitumens

New HDPE plastic bag (purpose to minimize impurities) was crushed in to small part and prepared 5% w/w of bitumen. Secondly, lignin was prepared 0.5% w/w of bitumen, respectively and then bitumen 60/70 was modified with 5% HDPE and modified lignin with those variables. Bitumen modification was performed at 160°C in 30 min using Hot Melt Mixing apparatus shown in Figure 1.

### 2.3. Characterizations

HDPE-modified bitumen will go through for characterization, FE-SEM images were taken at an



**Figure 1.** Hot Melt Mixing.

accelerated voltage of 20 kV, FTIR (Fourier Transform Infrared) were recorded using a Perkin-Elmer FTIR spectrometer, and each sample recording consisted of 30 scans recorded from 500 to 4000 cm<sup>-1</sup>. The ductility test of high density polyethylene (HDPE)-modified bitumen was tested based on ASTM D113.

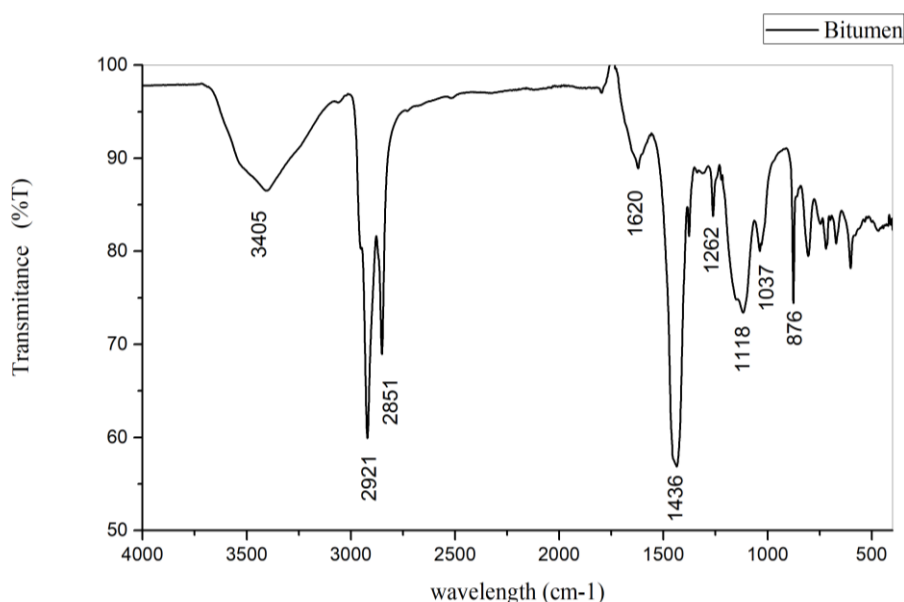
## 3. Result and Discussion

### 3.1. Base Material Characteristics

HDPE, bitumen, and modified lignin consist of certain compounds which give their own identity. Compounds are pure and homogeneous substances which consist of 2 or more different elements with certain comparisons that produce different properties from the constituent elements. The compound has a specific functional group which signifies its type. The functional group contained in a compound can

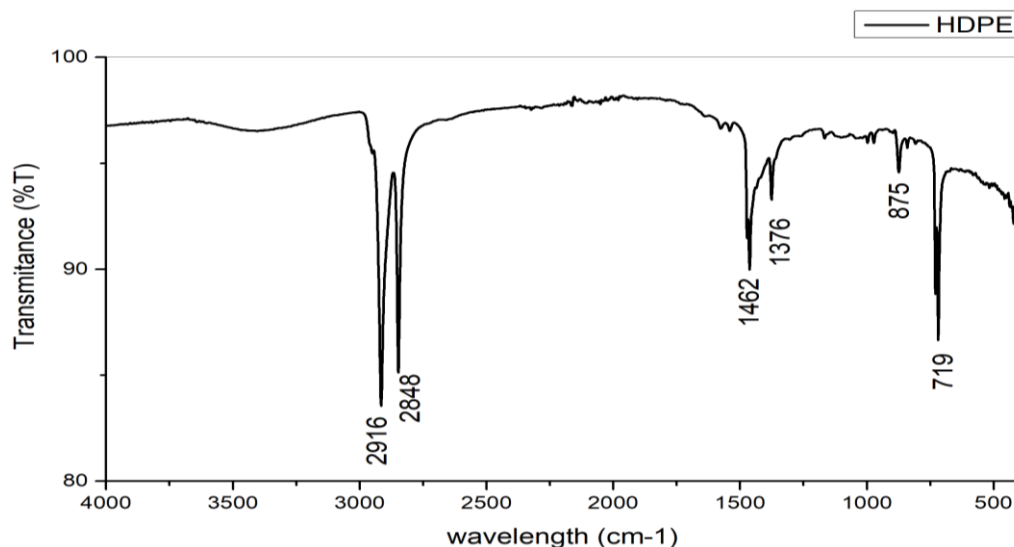
be known by FTIR test. Modified lignin characteristics could be found in our laboratory previous research [7].

Bitumen is a basic ingredient of a mixture which is a hydrocarbon compound which has a functional group  $-CH_3$  which is an aromatic lime substituent on bitumen,  $C-H$  which is the deformation of  $-CH_2$  and  $-CH_3$ , double chains  $C=O$  and  $C=C$ . From the graph in figure 2, there are several peaks that are produced from pure bitumen FTIR testing that is  $876\text{ cm}^{-1}$ ,  $1037\text{ cm}^{-1}$ ,  $1118\text{ cm}^{-1}$ ,  $1262\text{ cm}^{-1}$ ,  $1436\text{ cm}^{-1}$ ,  $1620\text{ cm}^{-1}$ ,  $2851\text{ cm}^{-1}$ ,  $2921\text{ cm}^{-1}$ , and  $3405\text{ cm}^{-1}$  where each peak shows a functional group. Peak widths at wavelengths between  $3050\text{--}3700$  indicate that the  $O-H$  bond is in the asphaltenes bitumen. Similar results were obtained by Brown [8]. The peak at  $2850\text{--}3000\text{ cm}^{-1}$  shows the presence of  $C-H$  alkane bonds in the main chain of bitumen. Furthermore, the peak which has a wavelength of  $1620\text{ cm}^{-1}$  indicates the existence of a  $C=C$  bond of an aromatic group. At peaks between  $1350\text{--}1400\text{ cm}^{-1}$  shows the presence of aromatic  $C-H$  bonds. Similar findings were reported by Xiangdao *et al.* [9].



**Figure 2.** FTIR of Bitumen.

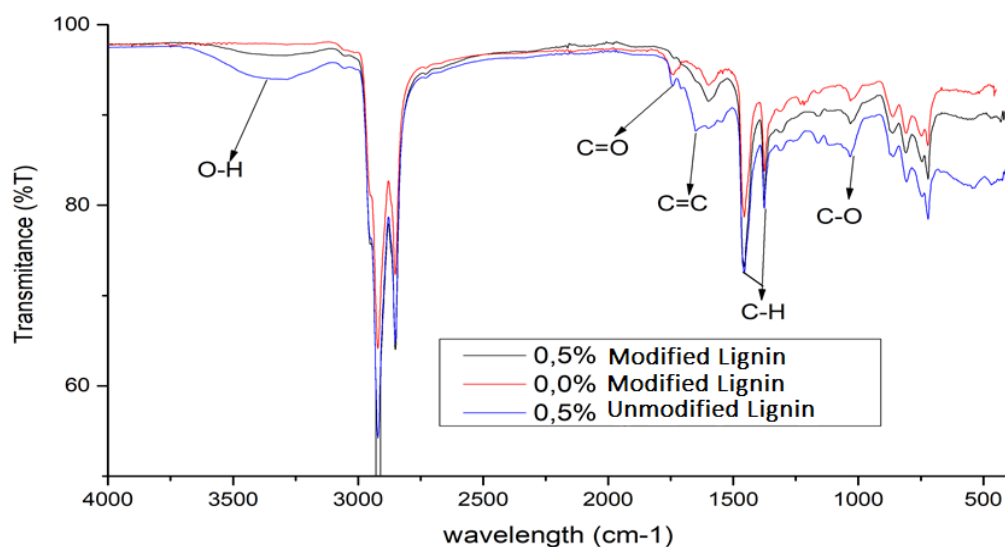
Addition of HDPE into the bitumen was aimed to contribute to the mechanical properties of the mixture to be better than the mechanical properties of pure bitumen so that the properties of the mixture have better mechanical strength than pure bitumen. This is because, the nature of HDPE which is harder than bitumen makes the results of the mixture tend to be more rigid than before it was given HDPE filler. Figure 3 have five peaks, namely  $2916\text{ cm}^{-1}$ ,  $2848\text{ cm}^{-1}$ ,  $1424\text{ cm}^{-1}$ ,  $874\text{ cm}^{-1}$ , and  $720\text{ cm}^{-1}$ . This shows that there is a  $C-H$  functional group which is an alkane bond with different deformations. Especially at the peak of  $2916\text{ cm}^{-1}$  and  $2848\text{ cm}^{-1}$  are asymmetric  $C-H$  and symmetric  $C-H$  which show HDPE identity. Similar tendency also was reported by Gulmine *et al.* [10].



**Figure 3.** FTIR of HDPE.

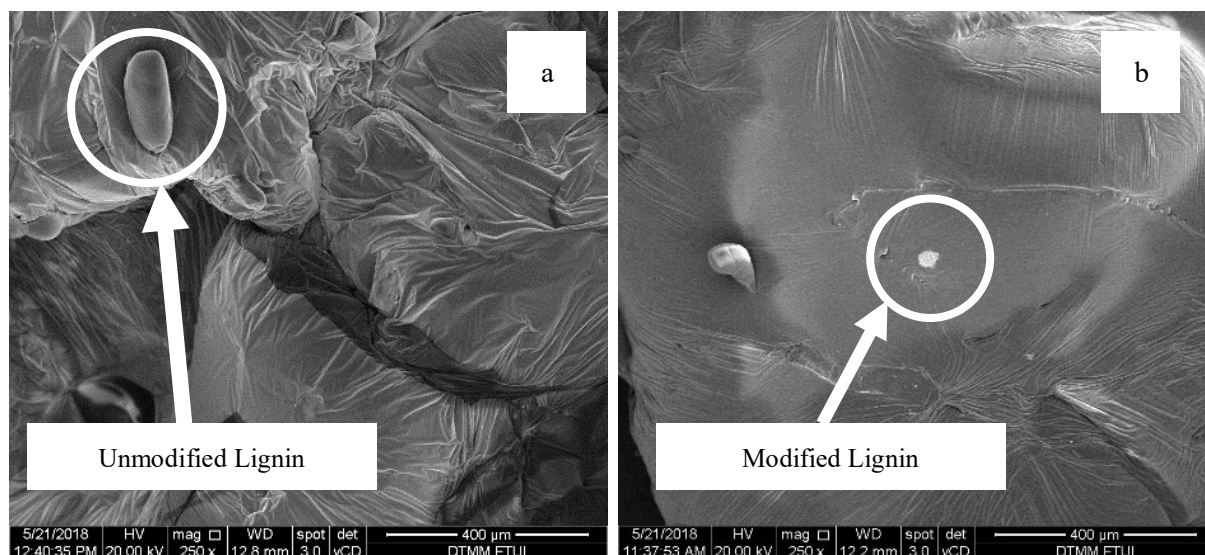
### 3.2. Compatibility of Modified Lignin

The compatibility discussion of modified bitumen, HDPE, and lignin mixture aims to see the effect of modified lignin concentration as a coupling agent so that we can find out the optimal modification of modified lignin. The role of coupling agent is a bridge that will form a chemical bond between bitumen and HDPE so that bitumen and HDPE plastic have good compatibility. FT-IR testing on the mixture aims to see whether a new functional group is formed between bitumen, HDPE, and modified lignin so that the mixture can be said to have good compatibility or not. From Figure 4 it can be compared to the functional groups owned by each mixture with a concentration of 5% wt lignin without modification, modified lignin 0% wt, and 5% wt. The visible peaks of PMB use lignin, without modified lignin, and using modified lignin is almost the same but the intensity is different.



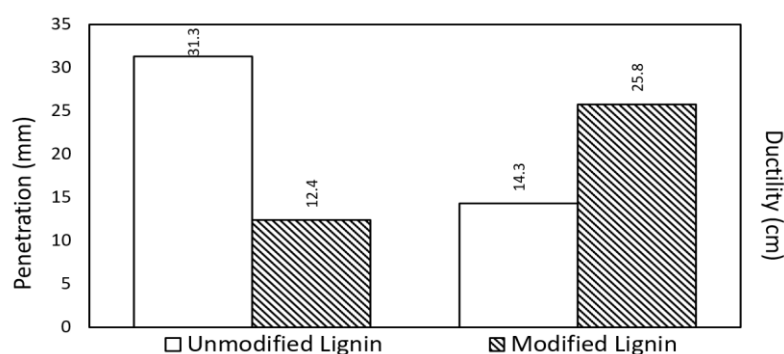
**Figure 4.** FTIR of PMB HDPE without lignin, with lignin and modified lignin.

Figure 4 indicated PMB HDPE with modified lignin compared to 0% wt lignin modification and 0.5 wt lignin at a peak of 2921  $\text{cm}^{-1}$  intensity that increased dramatically indicates the presence of CH<sub>2</sub> bond. In the fingerprint area and 3200-3500  $\text{cm}^{-1}$  shows the hydroxyl group (-OH) which is hydrophilic 0.5% wt modified lignin is between 0.5 wt lignin and 0% wt lignin modification. This shows that modified lignin has higher hydrophilic properties than 0% wt lignin and is lower than 0.5% lignin without modification and its hydrophobic properties are increased compared to 0% wt lignin modification and 0.5% lignin without modification.



**Figure 5.** Fracture Surface of PMB HDPE with (a) unmodified lignin and (b) modified lignin as a coupling agent.

Figure 5 showed that PMB HDPE between unmodified lignin and modified lignin shows a drastic difference in fracture surface in the same magnification. In the modified PMB HDPE lignin, the flat fracture surface indicates a strong bond on the bitumen interface and the modified lignin is perfectly attached. Whereas, PMB HDPE with unmodified lignin shows uneven fracture surface shape. The surface shape shows that the unmodified lignin does not adhere perfectly to the PMB HDPE, indicated weaker interface strength between bitumen and unmodified lignin. This shows that modified lignin has compatibility with bitumen and HDPE which is better than lignin without modification. The result of ductility test PMB HDPE using modified lignin is 25,8 cm while using unmodified lignin is 14,3 cm and the penetration test PMB HDPE using modified lignin is 12,4 mm while using unmodified lignin is 31,3 mm showed in figure 6, thus it furthermore indicated the compatibility is better in PMB HDPE using modified lignin than PMB HDPE using unmodified lignin.



**Figure 6.** Comparison of PMB HDPE with modified lignin and unmodified lignin.

#### 4. Conclusion

Modified lignin can be used as a coupling agent between the HDPE with bitumen which was indicated by the formation of ether peak and the loss of hydroxide peak better than unmodified lignin. FE-SEM result indicated that addition of modified lignin compared to unmodified lignin produce a better mixture between HDPE and bitumen. In addition, penetration value is lower and ductility value is higher in PMB HDPE with modified showed that the compatibility of the bitumen and HDPE with modified lignin was better than unmodified lignin.

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#### Acknowledgement

We appreciate and thank for the support from DRPM UI through *The Research Grant Scheme Program of PIT 9-2019* which makes this research possible.