



Preliminary Study on Functional Groups Characteristics of Asphalt Containing Rice Husk Silica

Simon Sembiring¹, Laili², Endah³, Suwarni⁴, Wasinton Simanjuntak⁵, Rudy Situmeang⁶

^{1,2,3,4} Department of Physics, Faculty of Mathematics and Natural Science, Universitas Lampung, Bandar Lampung 35145, Indonesia

^{5,6}Department of Chemistry Faculty of Mathematics and Natural Science, Universitas Lampung, Bandar Lampung 35145, Indonesia

Abstract. In this research, asphalt composite was produced by combining asphalt and rice husk silica. The ratio of asphalt and silica were 1:1.5; 1:1.7; and 1:1.9 respectively and calcined at temperature of 150°C. Functional group characteristics of asphalt composites were examined by FTIR and XRD. The FTIR and XRD studies revealed that the main Functional groups are Si-O-Si, C-H, and structure amorph of silica.

Keyword: Rice Husk, Silica, Asphalt, Functional Group, FTIR

Received 12 December 2018 | Revised [10 February 2019] | Accepted [28 February 2019]

1 Introduction

Asphalt has been widely used for roofs, sidewalks and sealing applications because of its good waterproof and binding properties. In the house roof industry, asphalt generally requires an air blowing process to reach the right softening and penetration point. Asphalt possesses a complex chemical composition that shows viscous and elastic properties which are very dependent on time and temperature. Previously, several studies had been conducted to modify and improve the characteristics of asphalt. For example, with the addition of other particles, asphalt cohesion and viscosity were reported to increase, which is efficient for high temperature conditions [1]. Some materials which are potential to be used to modify asphalt, such as hydrated lime, plastic or polymerized powder.

One of the additional inorganic materials that have been used intensively to improve the properties of asphalt is silica. Silica has also received great attention by the asphalt road designer for preparing asphalt materials with desirable properties since their excellent stability, low cost, high surface area, chemical purity, strong adsorption, and good dispersion ability [2],

^{*}Corresponding author at: Jl. Prof. Soemantri Brojonegoro No.1 Bandar Lampung, 35145, Indonesia

E-mail address: simonsembiring2@gmail.com

Copyright © 2019 Published by Talenta Publisher, ISSN: 2656-0747 e-ISSN: 2656-0755 Journal Homepage: https://talenta.usu.ac.id/index.php/JoTP

Journal of Technomaterial Physics Vol. 1, No. 1, 2019 | 63 – 68

[3] and [4]. For instance, silica has been used previously to strengthen a mixture of cement [5] and to strengthen elastomers such as solutes [6]. Since silica materials are highly reactive to asphalt binders rather than conventional fillers, and the ability to spread silica particles into asphalt binders, nano polymer composites with desired performance can be prepared [7], [8] and [9] modified asphalt binders with silica at a content of 4% and 6%, by weight. It has been observed that the performance of aging, rutting, and fatigue cracks of nanosilica modification binders has been improved.

Based on several studies, one source of silica is rice husk ash for agricultural waste is known to contain a number of silica, which can be extracted by a relatively simple method to obtain high purity, and active silica. For example, Haslinawati et al [10], Rafieel, et al [11] and Ugheoke et al [12] concluded that pure silica could be extracted with an excellent purity of 94-98%, presented as amorphous and reactive. Plenty efforts have been performed to utilize silica rice husks to produce various silica-based materials, such as nanosilica production [13] and [14]. In our previous study, several ceramic materials derived from silica rice husks reported in the literature were borosilicate [15], carbosil [16], aluminosilicate [17], mullite [18].

This research is a preliminary study with the aim to study the formation of functional groups or asphalt functionalities based on silica rice with a variety of silica which is used as raw material for making rice husk composites for roofing materials instead of lightweight steel roofs with silica variations. The characteristics of asphalt composites studied were the formation of structures and functional groups (functionality) using The X-Ray Diffraction (XRD), and Fourier Transform Infrared Spectroscopy (FTIR).

2 Materials and Methods

2.1 Materials

The skin of rice husk used as a silica source was obtained from the local rice milling industry in Bandar Lampung Province, Indonesia. Asphalt was obtained from the Buton refinery, Southeast Sulawesi Province, Indonesia. KOH, HCl, and alcohol (C_2H_5OH) were purchased from Merck (kGaA, Damstadt, Germany).

2.2 The fabrication of silica powder from rice husk

Rice husk silica was made through alkaline extraction method, following the procedure reported in previous studies [18]. The obtained soles were acidified by adding droplets of HCl 5% solution until the conversion of sol into the gel was completed. The gel was dried in an oven at a temperature of 110°C for eight hours and then grinded into powder. The silica material in white powder (Figure 1a), the x-ray diffraction pattern of the siloxane of rice husk (Figure 1b) and the x-ray diffraction pattern of the asphalt binder (Figure 1c), synthetic amorphous silica, which are polymorphs of silicon dioxide, SiO₂, has been used to modify the asphalt binder. The diffraction pattern shown in Figure 1b indicates that the sample is essentially amorphous, marked by the presence of a broad peak at 2θ in the range of $20-30^{\circ}$, caused by amorphous silica. Figure 1c shows that the sample is also amorphous, proven by the presence of two broad peaks at 2θ in the range of 15-30° and 35-50° and resulted by amorphous hydrocarbons.

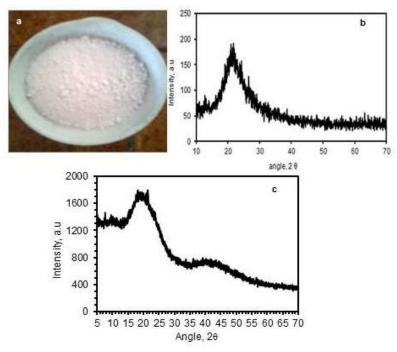


Figure 1. (a) Rice husk silica powder, (b) The XRD result of rice husk silica powder, and (c) The XRD result of asphalt binder

2.3 The manufacturing of asphalt composites

For the manufacture of asphalt composites, as much as 50 g of base asphalt binder was heated to 100°C and mixed with silica from rice husk using a shear mixer with a speed of 125 rpm for 1 hour. Rice husk Silica was mixed with base asphalt binder in a comparison of different asphalt and silica concentrations of (1.5; 1.7; and 1.9) to determine the effect of silica on the asphalt binder functional group.

2.4 Characterization

Furthermore, a spectroscopic analysis was conducted for it functions as a compound fingerprint. This was carried out on base asphalt binders using the infrared spectroscopy (FTIR), to assess the effectiveness of the addition of silica to the asphalt binder from the perspective of chemical functional groups. The infrared spectrum was recorded using the Perkin Elmer Fourier Transform Infrared (FTIR) spectrometer by scanning in wavenumbers ranging from 4000 to 400 cm⁻¹. The FTIR method is explained elsewhere [19]. All FTIR plots were recorded as a transmitted radiation against the wave numbers. The XRD pattern was collected using an automatic X-ray Shimadzu XD-610 diffractometer with Cuk α radiation ($\lambda = 0.15418$ Å, produced at 40 kV and 30 mA).

Figure 2a-c shows a spectrum of samples with a comparison of asphalt which is different from silica. A qualitative XRD was performed by comparing the diffraction line with the Standard Differentiation Diffraction method with the searching-matching method. Based on this method, the phase identified in the sample (Figure 2a-c) shows a large peak located around $2\theta = 22^{\circ}$, which indicates that silica is amorphous. Moreover, an amorphous carbon was discovered at $2\theta = 5-10^{\circ}$. The XRD pattern of the sample (Figure 2a) with the sample (Figure 2b-c), the significant effect of silica addition can be clearly observed.

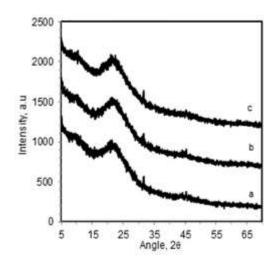


Figure 2. The X-Ray Diffraction pattern of sample with various ratio of asphalt and silica using the CuK α radiation, (a) 1: 1.5., (B) 1: 1.7, dan (c) 1: 1.9.

The FTIR spectrum from samples with different asphalt ratios to silica is illustrated in Figure 3a-c. FTIR was carried out to follow the reaction of sample dehydration and the formation of asphalt-silica bands. The results reveal the same functional group for all samples.

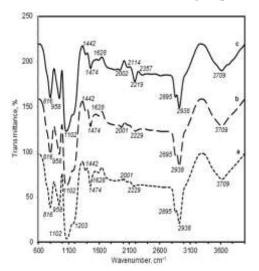


Figure 3. The FTIR Spectra of sample with various ratio of asphalt and silica (a) 1: 1.5, (b) 1:1.7, dan (c) 1: 1.9

shown in Figure 3 (a-c), the peak is located around 3709 cm⁻¹, which is usually assigned to the stretch vibration of the O-H bond. This absorption band most likely arises from O-H bonds in the silanol group in Si(OH)₄ and water molecules. The presence of water is confirmed by the absorption bands at 1628 cm⁻¹ [20], as a result of the bending vibrations of H-OH. The presence of Si(OH)₄ is supported by the absorption band at 1102 cm⁻¹, which usually corresponds to the stretch vibrations of Si-O-Si due to the deformation of Si-O bonds, as Tsai observed [21]. These findings are also in accordance with the previous research result [22], where it suggested that silica is intensively reactive to asphalt. The other strong bands were observed at wavelength of 2938 and 1474 cm⁻¹, which correspond to O-H stretching vibrations from carboxylic acids and C-H from alkanes. These findings are accordance to the results described in previous studies [23]. However, the addition of silica seems to affect the intensity of the asphalt chemical group; Therefore, it affects overall material performance. Thus, it can be concluded that the asphalt shows a change in bond for the addition of silica.

4 Conclusion

Based on a series of experiments carried out in this research, several conclusions were obtained regarding the analysis of the characteristics of asphalt composites with FTIR; namely (1) the formation of silica from rice husks with the detection of Si-OH, Si-O-Si, and CH from asphalt, (2) asphalt-silica formation consists of similar chemical functional groups and molecular structures, (3) The formation of asphalt composites was identified by CH and Si-O-Si functional groups.

Acknowledgement

Authors would like to thank and appreciate Ministry of Research, Technology and Higher Education of Republic of Indonesia for research funding provided via "Competence Research Grant Batch II No: 382/UN26.21/KU/2018.

REFERENCES

- [1] L. Ping and L. Yuniong, "Review on Nano Modified Asphalt," *Applied Mechanics & Materials*, vol. 587, pp. 1220-1223, 2014.
- [2] L. Senff, J. A. Labrincha, V. M. Ferreira, D. Hotza and W. L. Repette, "Effect of Nano Silica on Rheology and Fresh Properties of Cement Pastes and Mortars," *Construction Building Material*, vol. 23, no. 7, pp. 2487-2491, 2009.
- [3] D. Kong, X. Du, S. Wei, H. Zhang, Y. Yang and S. P. Shah, "Influence of Nano-silica Agglomeration on Microstructure and Properties of the Hardened Cement Based Materials," *Construction Building Material*, vol. 37, pp. 707-715, 2012.
- [4] L. P. Singh, S. R. Karade, S. K. Bhattacharyya, M. M. Yousuf and S. Ahalawat, "Beneficial role of nanosilica in cement based materials - a review," *Construction Building Material*, vol. 47, pp. 1069-1077, 2013.
- [5] G. Quercia and Brouwers, "Application of Nano-silica (nS) in Concrete Mixtures," 8th fib PhD Symposium in Kgs. Lyngby, Denmark, 2010.

- [6] K. Chrissafis, K. M. Paraskevopoulos, G. Z. Papageorgio and D. N. Bikiaris, "Thermal and Dynamic Mechanical Behavior of Bionanocomposites: Fumed Silica Nano Particles Dispersed in Poly (vinyl pyrrolidone), Chitosan, and Poly(vinyl alcohol)," *Journal of Applied Polymer Science*, vol. 110, pp. 1739-1749, 2008.
- [7] P. C. Lebaron, Z. Wang and T. J. Pinnavaia, "Polymer-layered Silicate Nanocomposites: an Overview," *Applied Clay Science*, vol. 15, no. 1-2, p. 11–29, 1999.
- [8] S. S. Ray and M. Okamoto, "Polymer/layered Silicate Nanocomposites: a Review from Preparation to Processing," *Program Polymer. Science*, vol. 28, no. 11, p. 1539–1641, 2003.
- [9] H. yao, Z. You, L. Li, C. H. Lee, D. WIngard, Y. K. Yap, X. Shi and S. W. Goh, "Rheological Properties and Chemical Bonding of Asphalt Modified with Nanosilica," *Journal Material Civil Engineering*, vol. 25, no. 11, pp. 1619-1630, 2012.
- [10] M. M. Haslinawati, K. A. Matori, Z. A. Wahab, H. A. A. Sidek and A. T. Zainal, "Effect of Temperature on Ceramic from Rice Husk Ash," *International Journal of Basic & Applied Sciences*, vol. 09, no. 09, pp. 22-25, 2009.
- [11] E. Rafieel, S. Shahebrahimi, M. Feyzil and M. Shaterzadeh, "Optimization of Synthesis and Characterization of Nanosilica Produced from Rice Husk a Common Waste Material," *International Nano Letters*, vol. 2, no. 29, pp. 1-8, 2012.
- [12] B. I. Ugheoke and O. Mamat, "A Novel Method for High Volume Production of Nano silica from Rice Husk: Process Development and Product Characteristics," *International of Materials Engineering Innovation*, vol. 3, no. 2, pp. 139-155, 2012.
- [13] K. Amutha, R. Ravibaskar and G. Sivakumar, "Extraction, Synthesis and Characterization of Nanosilica from Rice Husk Ash," *International Journal of nano Technology and Applications*, vol. 4, no. 1, pp. 61-66, 2010.
- [14] T. H. Liou and C. C. Yang, "Synthesis and Surface Characteristics of Nanosilica Produced from Alkali-extracted Rice Husk Ash," *Material Science Engineering B*, vol. 176, p. 521– 529, 2011.
- [15] S. Sembiring, "Synthesis and Characterisation of Rice Husk Silica Based Borosilicate (B2SiO5) Ceramic by Sol Gel Routes," *Indonesian Journal of Chemistry*, vol. 11, no. 1, pp. 85-89, 2011.
- [16] W. Simanjuntak, S. Sembiring and K. Sebayang, "Effect of Pyrolysis Temperature on Composition and Electrical Conductivity of Carbosil Prepared from Rice Husk," *Indonesian Journal of Chemistry*, vol. 12, no. 1, pp. 119-125, 2012.
- [17] W. Simanjuntak, S. Sembiring, P. Manurung, R. Situmeang and I. M. Low, "Characteristics of Aluminosilicates Prepared from Rice Husk Silica and Aluminum Metal," *Ceramic International*, vol. 39, no. 8, pp. 9369-9375, 2013.
- [18] S. Sembiring, A. Riyanto, W. Simanjuntak and R. Situmorang, "Effect of MgO-SiO2 Ratio on the Forsterite (Mg2SiO4) Precursors Characteristics Derived from Amorphous Rice Husk Silica," *Journal Oriental Chemistry*, vol. 33, no. 4, pp. 186-192, 2017.
- [19] S. S. Karahancer, M. Kiristi, S. Terzi, M. Saltan, A. U. Oksuz and L. Oksuz, "Performance Evaluation of Nano-modified Asphalt Concrete," *Construction Building Material*, vol. 71, pp. 283-288, 2014.
- [20] F. Adam and J. H. Chua, "The Adsorption of Palmytic Acid on Rice Husk Ash Chemically Modified with Al (III) Ion Using the Sol-gel Technique," *Journal of Colloid and Interface Science*, vol. 280, no. 1, pp. 55-61, 2004.
- [21] M. T. Tsai, "Effect of Hydrolysis Processing on the Character of Forsterite Gel Fibers Part II:Crystallites and Microstructural Evolution," *Journal of European Ceramic Society*, vol. 22, pp. 1085-1094, 2002.
- [22] F. Zafari, M. Rahi, N. Moshtagh and H. Nazockdast, "The Improvment of Bitumen Properties by Adding Nanosilica," *Study of Civil Engineering and Architecture (SCEA)*, vol. 3, pp. 62-69, 2014.
- [23] M. Enieb and A. Diab, "Characteristics of Asphalt Binder and Mixture Containing Nanosilica," *International Journal of Pavement Research and Technology*, vol. 10, pp. 148-157, 2017.