

Waste Cooking Oil as Bio Asphalt Binder: A Critical Review

P J Ramadhansyah¹, K A Masri¹, W N A Wan Azahar², N Mashros³, A H Norhidayah³, M N Mohd Warid³, M K I M Satar³ and H Yaacob³

¹Faculty of Civil Engineering Technology, Universiti Malaysia Pahang, 26300 Gambang, Pahang, Malaysia

²Kulliyah of Engineering, Department of Civil Engineering, International Islamic University Malaysia, 50728 Kuala Lumpur, Malaysia

³Faculty of Engineering, School of Civil Engineering, Universiti Teknologi Malaysia, 81310 Johor Bahru, Malaysia

Abstract. The modification of binder with waste cooking oil (WCO) for paving materials indicates the implementation of recycling practice with environmental issues concern, thus improving the proper management of this waste product. This paper presents a critical review of the WCO utilisation as a modifier to enhance binder properties. The review was focuses on the evaluation of WCO in asphalt binder modification and asphaltic concrete mixture. Basically, oil-based modification by using WCO in paving material provides a promising waste material potential in improving the engineering value in terms of rheological and mechanical performance for modified binder and asphalt mixture. It is expected that the addition of this modifier in binder gives superior performance and is comparable with the conventional binder.

Content

1. Introduction	1
1.1. Asphalt Binder Modification	2
2. Waste Cooking Oil.....	2
2.1. Environmental issues.....	2
2.2. Chemical characteristic of WCO	3
2.3. Physiochemical Properties.....	4
3. Recyclability of WCO as Bio- Binder.....	5
3.1. WCO in Pavement.....	7
4. Discussions.....	8
4.1. Proposed solution.....	8
5. Conclusions.....	9
References.....	9
Acknowledgements.....	11

1. Introduction

Waste materials are widely used as a constituent part or a substitution element in the production of modified asphalt binder to modify conventional binder properties. This waste material is obtained from household, commercial, and industrial by-products. In fact, these waste products have a discreet and specific property, which enable them to be an additive or a modifier element. These additives come in liquid and powder forms that are desirable for modifying and upgrading construction material properties. In order to prevent pavement distress and minimise the total maintenance work cost, the bituminous layer should be strengthened with high quality recycled waste materials to improve its

mechanical properties in terms of permanent deformation resistance, fatigue and so on. The application of waste and recycled product has fulfilled the sustainable development requirement so that a green highway can also be achieved.

1.1. Asphalt Binder Modification

An effort for binder performance improvement has induced the evaluation, development and use of various asphalt binder modifiers. Currently, a number of low-cost asphalt modification and extension technique is proposed to minimise the fluctuating cost of binder production, while at the same time enhance the asphalt binder performance. Modified asphalt offers properties enhancement in mitigation of pavement distress and reduces life cycle costs when compared to the conventional binder. Besides, in addressing various arising issues regarding pavement performance, therefore asphalt binder modification is suggested as one effective technical solution to solve the problem faced by asphalt industry [1]. According to Hainin et al. [2], binder properties alteration and improvement can be achieved with modified asphalt binder which in turn ensures a long-term pavement performance. Due to this fact, many researches were carried out to investigate the potential application of waste materials in the construction industry. Basically, a number of noteworthy worldwide researches were conducted in applying a modifier from waste oil source. In recent years, a wide range of oil-based modifications was introduced in the asphalt industries to achieve some desirable properties in material for pavement construction. The oil-based source can be categorised as bio-oil from biomass production industry, waste engine oil from automotive industry and waste cooking oil from agricultural industry. Based on these oil types, numerous researches have explored the potential of modifiers from biomass supply [3]. Meanwhile some were focused on waste oil product, such as waste cooking oil (WCO) [4]. Due to its high availability and low cost feedstock, WCO source has become intensively applied in asphalt binder modification. Most previous research has recorded WCO application for the conventional bitumen replacement, bitumen extender and modifier [5].

2. Waste Cooking Oil

Waste cooking oil is non-edible oil and can be recognised as a by-product of fresh oil produced during food frying [6] as shown in Figure 1. Fresh cooking oil is physically light yellow in color with low viscosity, while used cooking oil has high viscosity and dark brown in color [7]. WCO source is collected from food industries, household disposables, restaurants and recycling centre by authorised companies [8]. The annual WCO collection is approximately three billion gallons [9]. Meanwhile, according to Math et al. [10] about 15 million tonnes of WCO production per year was recorded based on statistics. In Malaysia approximately 0.5 million tonnes of WCO is generated per year and derived from palm oil source. An increase in demand for food consumption (Phan and Phan, 2008), besides rapid and tremendous growth of human population [11] is identified as the main factor that attributed to the huge amount of WCO production.

2.1. Environmental issues

At present, the WCO recycling is at an empirical stage, in which only a small amount of used cooking oil is properly collected and recycled especially from residential area and food restaurants. In fact, enormous WCO quantity, without any appropriate treatment, is illegally dumped into landfills and rivers which have caused serious adverse environmental impact, especially environmental pollution issues. The most common environmental issue that arise is the eutrophication process, whereby this phenomenon is caused by the blockage of sunlight from passing through the river surface because of the presence of a thin layer of oil. Apart from sunlight, oxygen supply to aquatic life is also disrupted by this process [12], thus leading to the excessive growth of micro-organisms, phytoplanktons and algae. River water quality is measured by the existence of these micro-organisms wherein their huge growth rate affects the intrinsic equilibrium of aquatic ecosystem. In residential area community, clogging problem has grabbed other human's attention because of blocked drains. This blockage happens when waste frying oil is poured into the kitchen sink and accumulates in the drains. This issue has affected waste water treatment plants and indirectly maximised the processing costs since routine maintenance work must be done in order to mitigate and monitor this clogging problem. The

estimation of about 40% of sewerage system blockages are contributed by pouring of waste cooking oil into the kitchen sinks. In addition, the soil has high potential to be contaminated by waste frying oil as the eco-toxic possessed in this oil content can damage plants. Due to dumping problems and possible water and land resources pollution, supervision and management of waste cooking oil has created a significant challenge that needs consideration. Therefore, waste cooking oil recycling is becoming an effective option to mitigate pollution issues due to illegal dumping problem with the greatest concern in high construction cost and natural resource conservation [13]. According to Alcantara et al. [14], the mitigation of major environmental issues, such as waste oil disposal and waste water treatment, can be encountered by the recyclability of WCO as a modifier in asphalt binder modification.



Figure 1: Waste cooking oil sample after frying process

2.2. Chemical characteristic of WCO

The majority of study stated that the predominant compounds found in WCO are Oleic acid, Palmitic acid and Linoleic acid as listed in Table 1. For example, Udomsap et al. [15] and Wen et al. [16] reported that the major components of WCO are Linoleic acid, followed by Oleic acid and Palmitic acid. Meanwhile, Ma and Hanna [17] discovered that the dominant component found in WCO is Palmitic acid, Oleic acid and Linoleic acid. Basically, Lauric acid, Myristic acid, Palmitic acid and Stearic acid are recognised as the saturated fatty acids, while Linoleic acid, γ Linolenic acid and Linolenic acid are identified as polyunsaturated fatty acids [15]. Oleic acid and Cis-11-Eicosenoic are monounsaturated fatty acids. The saturated component can be depicted as the non-existence double-bond in WCO fatty acids. This implies that maximum number of possible hydrogen atoms per atom carbon is contained in these chains [18]. Otherwise, the presence of double-bond in unsaturated component indicates insufficient maximum number of possible hydrogen atoms, thereby will induce the electronegativity to react during chemical reaction in which this unsaturated component tend to accept the hydrogen atoms.

Table 1: Free fatty acids in waste cooking oil

Fatty Acids	Chemical Formula	Waste Cooking Oil (%)		
		Ma and Hanna [17]	Udomsap <i>et al.</i> [15]	Wen <i>et al.</i> [16]
Palmitic acid	C16:0	42.8	10.2	8.5
Oleic acid	C18:1n9c	40.5	22.8	21.2
Linoleic acid	C18:2n6c	10.1	53.7	55.2

The different fatty acids composition is recorded if different sources of waste cooking oil are used as tabulated in Table 2. These acids are categorised as Free Fatty Acids (FFA) that is naturally contained in fats, oils and greases, in which higher FFA component in WCO leads to higher acid value [19]. Theoretically, the fatty acids chain and number of double bonds affect oil physical and chemical properties [20] since the degree of unsaturated compound is more preferable for chemical reactions. The acid composition in WCO can be categorised as saturated, monounsaturated and polyunsaturated. The comparison of FFA composition between fresh cooking oil and waste cooking oil reported by Maneerung et al. [21] indicated that the reduction of unsaturated fatty acids consists of Oleic acid, Linoleic acid and Linolenic acid in WCO after frying process, as shown in Table 2. The decreasing trends of unsaturated fatty acids are attributed to the chemical degradation reaction during frying in which these acids tend for hydrogen acceptance thereby increases the saturated fatty acids.

Table 2: FFA composition in fresh and waste cooking oil [21]

Free Fatty Acid Composition (%)	Fresh Cooking Oil	Waste Cooking Oil
Oleic (C ₁₈ H ₃₄ O ₂)	43.9	43.2
Linoleic (C ₁₈ H ₃₂ O ₂)	30.4	30.1
Palmitic (C ₁₆ H ₃₂ O ₂)	20.3	19.4
Linolenic (C ₁₈ H ₃₀ O ₂)	4.8	4.7
Stearic (C ₁₈ H ₃₆ O ₂)	2.9	2.6

2.3. Physiochemical Properties

Ullah et al. [22] conducted test properties to compare the properties of unused and used cooking oil as presented in Table 3. The various properties test conducted were acid value, calorific value, peroxide value, density, kinematic viscosity, dynamic viscosity, flash point and moisture content. All these properties were measured to identify the WCO quality. The main parameter for physiochemical properties was evaluated based on the acid value test, density and viscosity. The acid value, density and viscosity (dynamic and kinematic) result obtained indicates an increment in value from unused to used cooking oil. Increasing acid value represents a higher value of free fatty acid content in WCO as these properties are related to each other. According to Chhetri et al. [23], the free fatty acids contained in waste cooking oil are higher as compared to fresh cooking oil. These findings coincide with Choe and Min [6], in which deep fat frying causes parameter alteration, where an increase in free fatty acids content, density and viscosity was observed in WCO. In addition, the density increase is attributed to the decrease in unsaturated fatty acids because the chemical reaction during frying oil has changed into saturated fatty acids. This can be observed by the physical WCO appearance wherein the unused cooking oil is light in condition while used cooking oil is thicker and more viscous. During frying, the oil will undergo oil polymerisation, producing cyclic monomer and polymer. This process depends on the degree of unsaturation, in which the increase of cyclic compound formation is recorded as increase in the linolenic acid, since these types of acid is categorised as unsaturated fatty acid. The polymer compound produced from the polymerisation accelerates oil degradation, thereby will increase the WCO viscosity [24]. Besides, the other factor that attributed to viscosity value is increase in polar formation and oxygen containing molecule. Therefore, it can be noticed that the used cooking oil viscosity is higher in comparison with unused cooking oil.

Table 3: Physiochemical properties of unused and used cooking oil [22]

Properties	Unused cooking oil values	Used cooking oil values
------------	---------------------------	-------------------------

Acid value (mg KOH/gm)	0.3	4.03
Calorific value (J/gm)	-----	39658
Peroxide value (meq/kg)	< 10	10
Density (gm/cm ³)	0.898	0.9013
Kinematic Viscosity (mm ² /s)	39.994	44.956
Dynamic Viscosity (mpa.s)	35.920	40.519
Flash point (°C)	161-164	222-224
Moisture content (wt %)	0.101	0.140

3. Recyclability of WCO as Bio-Binder

A modifier can be defined as a potential material which enhances the service life performance and can withstand the loading factor exposure for improvement of asphalt mixture engineering properties [1]. According to Arabani et al. [25], the substitution of a modifier as part of the modification mechanism in asphaltic concrete mixture has reduced the deterioration rate due to presence of asphalt pavement distress. Based on the listed previous research, WCO was mostly applied as a rejuvenator since the fluidity factor in WCO has the capability for restoration aged binder by reducing its viscosity. Apart from these, the modification of binder by using WCO as a modifier was focused. Basically, the application of a rejuvenator is desirable to determine the cracking resistance at low temperature, while the function of a modifier is preferable for rutting resistance at high temperature. Maharaj et al. [26] performed an assessment for rutting and cracking resistance performance of modified asphalt binder by incorporating WCO as a modifier. The two factors which attributed to the rheological performance, represented as WCO content and test temperature, were measured to assess the potential of these parameters affecting rutting and cracking performance. The rheological performance demonstrated that the decreasing $G^* \cdot \sin \delta$ was observed (see Figure 2) which indicated an improvement in fatigue cracking resistance. The increasing trend of fatigue cracking resistance was recorded as the WCO concentration was increased in the modified binder. In contrast, with the increasing WCO concentration in control binder, the $G^*/\sin \delta$ was decreased which explained the decrement of rutting resistance performance, as shown in Figure 3.

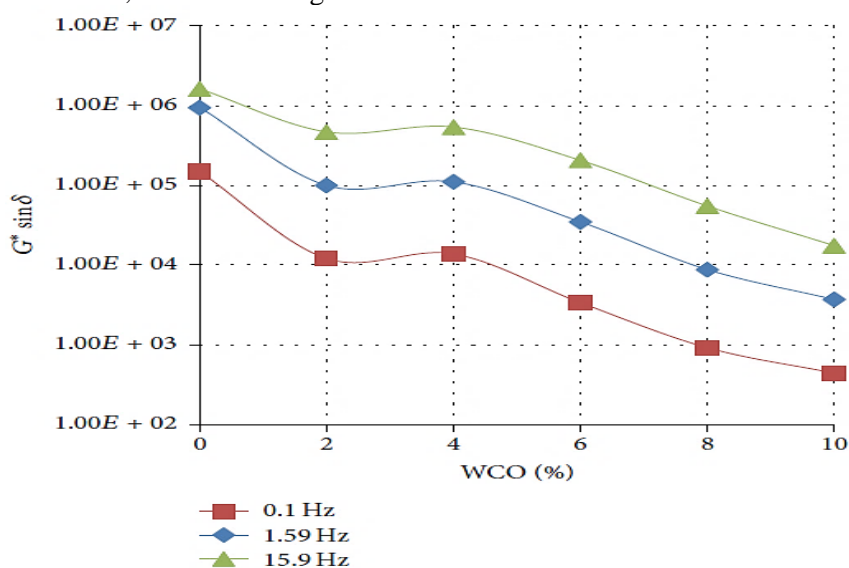


Figure 2: $G^* \cdot \sin \delta$ versus WCO percentage [26]

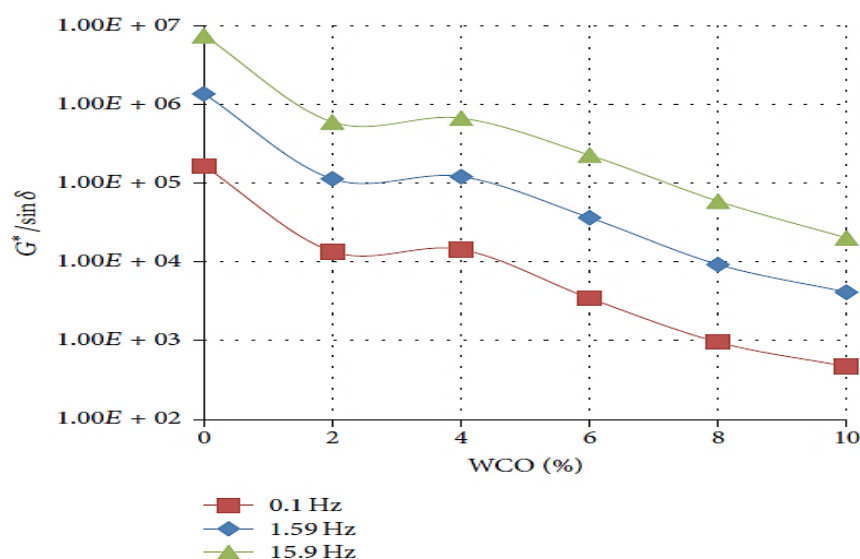


Figure 3: $G^*/\sin \delta$ versus WCO percentage [26]

Previously, Wen et al. [16] conducted a laboratory evaluation for binder containing bio-asphalt from the conversion of WCO through a thermochemical process. There were three different types of base binder used in this study which is PG 58-28, PG 76-22 and PG 82-16. About 30% and 60% of bio-asphalt was blended with PG 58-28 meanwhile the bio-asphalt was mixed at 10% and 30% (by the weight of total binder) for PG 76-22 and PG 82-16 binders. The rutting resistance performance achieved in this research coincided with the previous study, wherein increasing non-recoverable creep compliance (J_{nr}) value from multiple stress creep and recovery (MSCR) result was recorded with the addition of bio-asphalt as listed in Table 4. The increment of J_{nr} indicated a high susceptibility of modified binder with bio-asphalt to rutting exposure. Besides, the lower critical strain energy density (CSED) attained in this study resulted in the reduction of fatigue cracking resistance with the addition of bio-asphalt content in the conventional binder. The low value of CSED was attributed by the lower shear strength possessed in the modified binder with bio-asphalt as compared to the control binder. The rheological findings indicated that the modified binder with bio-asphalt reduced the resistance to permanent deformation and fatigue cracking.

Table 4: Non-recoverable creep compliance (J_{nr}) results [16]

PG	Bio-asphalt (%)	Stress level (Pa)	J_{nr} (1/Pa)
58-28	0	100	0.94
58-28	0	3,200	1.14
58-28	30	100	1.24
58-28	30	3,200	1.60
58-28	60	100	0.50
58-28	60	3,200	1.07
82-16	0	100	1.02
82-16	0	3,200	2.17
82-16	10	100	1.30
82-16	10	3,200	3.61
82-16	30	100	1.94
82-16	30	3,200	7.04
76-22	0	100	0.94

76-22	0	3,200	1.36
76-22	10	100	1.45
76-22	10	3,200	2.43
76-22	30	100	2.37
76-22	30	3,200	4.70

3.1. WCO in Pavement

A laboratory evaluation conducted by Wen et al. [16] reported that the addition of waste cooking oil based bio-asphalt to produce alternative binder for hot mix asphalt (HMA) reduced the dynamic modulus, wherein the stiffness was linearly decreased with the addition of bioasphalt concentration. The decrement of flow number with increasing amount of bioasphalt (see in Figure 4) also proved the reduction in rutting resistance performance, which is more prone to deflection than a control mixture. Another interpretation indicated that adding bio-asphalt reduces high-temperature performance as expected.

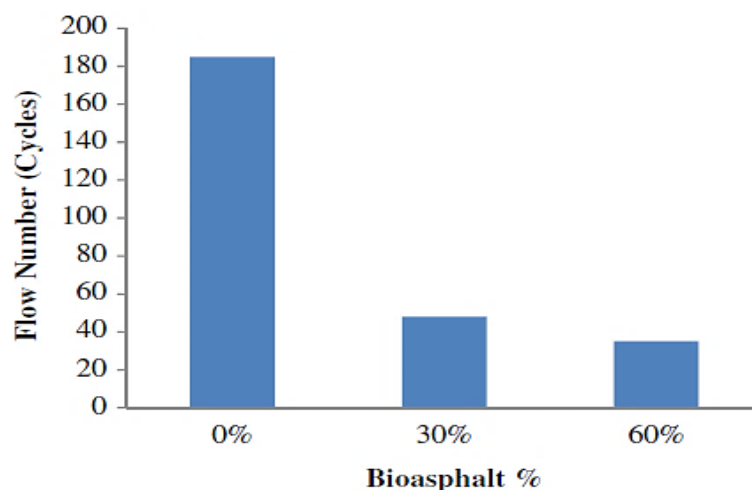


Figure 4: Flow number at different percentage of bio-asphalt from WCO [16]

Other investigation by Bailey and Zoorob [27] proved an opposite findings, where the addition of Used Vegetable Oil (UVO) has increased the Indirect Tensile Stiffness Modulus (ITSM) performance which implied that the modified asphalt mixture with UVO is more durable as compared to the control mixture. The laboratory result was validated with the field evaluation in which the wheel tracking test proved that the lowest rut depth was exhibited by the mixture incorporated with UVO relative to the control mixture. A similar research finding was reported by Some et al. [28], in which a lower rutting depth was observed for modified binder with oil derived from sunflower and rapeseed source (vegetable oil) in comparison with the control mixture. This implied that, an improvement in the resistance to permanent deformation was recorded with the addition of waste vegetable oil. The trend of the compression strength was predicted since a decreasing complex modulus was observed in the binder test [28].

4. Discussions

From the review presented, it can be noticed that WCO can be categorised as one waste material that is mostly utilised for partial substitution as a modifier in binder modification as reported by the previous research work. Oil-based modification by using WCO is expected to improve the engineering value of modified asphalt binder in comparison with the conventional binder. Apart from that, waste disposal problem could be solved and also promotes environmental preservation. A significant and noticeable finding reported by previous researchers indicating that the WCO application is recorded as a superior improvement for cracking resistance performance at low temperature. The superior

performance of the modified binder incorporating WCO is attributed to the natural fluidity characteristic of WCO. Theoretically, at low temperature exposure, bitumen undergoes high brittleness rate and is prone to experience cracking failure at this temperature. As a result, the presence of cracking increases the tendency for noticeable stress. The WCO fluidity properties in binder facilitates high flow rate and decreases the viscosity of modified binder within the mix, thereby assisting the reduction in induced stress or cracking existence. However, available information on the effect of modified binder containing WCO on rutting resistance at high temperature is very limited as the adverse effect is widely reported and still unresolved at the moment. Basically, the addition of WCO in binder causes the softening effect on the modified binder and it is expected that the soft binder contributes to poor performance in permanent deformation resistance due to unable to withstand with high temperature exposure. The justification is presently not well documented and there is no conclusion can be drawn in explaining the reason for this mechanism. Borhan et al. [29] justified the main factor that attributed to this issue where it is due to the incompatibility characteristic of the oil source and asphalt binder, thereby inducing instability problem in the chemical reaction between oil in modified binder and asphalt mixture. Despite that, this factor is not a broad concern of other research as WCO is applied directly in the binder without determining the WCO fundamental quality parameter that might affect the modified binder with WCO performance. Due to the arising issue, several valuable efforts were triggered in this study to determine WCO quality and investigate the compatibility properties between WCO and asphalt binder for the modified binder incorporating WCO improvement. Therefore, it is significant to conduct a chemical WCO quality test and perform chemical analysis to discover whether WCO is compatible with the binder for binder modification. Once the WCO quality and compatibility properties are determined, chemical pre-treatment is proposed in producing treated WCO with improved performance than untreated WCO. For this reason, it is vital to evaluate the effect of untreated and treated WCO on the binder and asphalt mixture performance after conducting the chemical treatment.

4.1. Proposed solution

The potential solution is proposed based on the improvement in certain properties, such as oxidative stability, free fatty acids content, low temperature and antioxidant content. One of the major challenges when dealing with oil is low oxidative stability which requires improvement to mitigate the problem. In combating poor oxidative stability performance, three types of solution were proposed, such as genetic modification, chemical modification and various additives with antioxidant materials usage. Genetic modification is explored by the soybean seed alteration and suggested by producing the low linolenic acid which is recognised as one potential alternative solution for hydrogenation (Mounts et al., 1994). The mechanism for chemical modification is conducted by shifting the fatty acids component in oil. The chemical modification through the trans-esterification process is proposed for minimising the high free fatty acids content in oil [30].

5. Conclusions

The significant noticeable findings indicated that WCO application has recorded the greatest improvement in cracking resistance performance at low temperature. Contrast, an adverse effect was observed on high temperature for rutting resistance performance. However, many studies were conducted and are still being developed to discover more about the WCO potential in modified asphalt binder performance, besides finding another alternative solution to enhance rutting resistance performance. Nevertheless, there is still a long journey to go through before the proposed modifier is widely accepted and broadly utilised in pavement industry. More expanding research work is required in future to prove the superiority of WCO application in pavement material.

6. References

- [1] Teymourpour P, Sillamäe S and Bahia H U 2015 *Road Mater. Pavement*. **16** pp. 50-74.
- [2] Hainin M R, Warid M N M, Izzul R, Ruzaini M K and Yusak M I M 2014 *Adv. Mater. Res.* **911** pp. 449-453.
- [3] Chen M Xiao F, Putman B, Leng B and Wu S 2014 *Constr. Build. Mater.* **59** pp. 10-16.

- [4] Yang X, You Z, Dai Q and Mills-Beale J *Constr. Build. Mater.* **51** pp. 424–431.
- [5] Ramadhansyah P J, Azman M K, Idris A M, Nur Aifa W A W, Ekarizan S, Hainin M R, Norhidayah A H and Haryati Y 2019 *IOP Conf. Series.: Earth. Environ. Sci.* **244**(1) pp. 1-6.
- [6] Choe E and Min D B 2007 *J. Food Sci.* **72**(5) pp. 1–10.
- [7] Singhabhandhu A and Tezuka T 2010 *Energy.* **35**(6) pp. 2544–2551.
- [8] Zhang H, Wang Q and Mortimer S R 2012 *Renew. Sust. Energ. Rev.* **16**(7) pp. 5225–5231.
- [9] Putra Jaya R, Masri K A, Awang H, Ali M I, Ramli N I, Wan Azahar W N A, Shaffie E, Yaacob H, Mohd Warid M N and Ramli 2019 *Int. J. Recent Technol. Eng.* **7**(6) pp. 16-19.
- [10] Math M C, Kumar S P and Chetty S V 2010 *Energy. Sustain. Dev.* **14**(4), 339–345.
- [11] Chen Y, Xiao B, Chang J, Fu Y, Lv P and Wang X 2009 *Energy Convers. Manag.* **50**(3) pp. 668-673.
- [12] Hamad B S, Rteil A A and El-fadel M 2003 *Constr. Build. Mater.* **17** pp. 311–318.
- [13] El-Fadel M and Houry R 2001 *Resour. Conserv. Recy.* **33** pp. 75–91.
- [14] Alcantara R, Amores J, Canoira L, Fidalgo E, Franco M J and Navarro A 2000 *Biomass Bioenergy.* **18** pp. 515–527.
- [15] Udomsap P, Chollacoop N, Topaiboul S and Hirotsu T 2009 *I. J. Renew. Energ.* **4**(2) pp. 47-59.
- [16] Wen Z, Yu X, Tu S T, Yan J and Dahlquist E 2010 *Bioresour. Technol.* **101** pp. 9570-9576.
- [17] Ma F and Hanna M A 1999 *Bioresour. Technol.* **70** pp. 1-15.
- [18] Lam M K, Lee K T and Mohamed A R 2010 *Biotechnol. Adv.* **28** pp. 500-518.
- [19] Silitonga A S, Masjuki H H, Mahlia T M I, Ong H C, Atabani A E and Chong W T 2013 *Renew. Sust. Energ. Rev.* **24** pp. 514–533.
- [20] Zlatanic A, Lava C, Zhang W and Petrovic Z S 2004 *J. Polym. Sci. Pol. Phys.* **42**(5) pp. 809–819.
- [21] Maneerung T, Kawi S, Dai Y and Wang C H 2016 *Energy Convers. Manag.* **123** pp. 487–497.
- [22] Ullah Z, Bustam M A and Man Z 2014 *Int. J. Chem. Eng.* **5**(2) pp. 134-137.
- [23] Chhetri A B, Watts K C and Islam M R 2008 *Energies.* **1**(1) pp. 3-18.
- [24] Tseng Y C, Moreira R G and Sun X 1996 *J. Food. Sci. Technology.* **31** pp. 287– 294.
- [25] Arabani M, Mirabdolazimi S M and Sasani A R 2010 *Constr. Build. Mater.* **24** pp. 1060-1068.
- [26] Maharaj R, Harry V R and Mohamed N 2015 *Sci. World. J.* **15** pp. 1-7.
- [27] Bailey H K and Zoorob S E 2012 *5th Eurasphalt - Eurobitume Congress.* Pp. 1-12, Istanbul.
- [28] Some S C, Pavoine A and Chailleux E 2016 *Int. J. Pavement Res. and Technol.* **9** pp.368-375.
- [29] Borhan M N, Suja F, Ismail A and Rahmat R A O K 2009 *Eur. J. Sci. Res.* **28**(3) pp. 398-411.
- [30] Phan A N and Phan T M 2008 *Fuel.* **87** pp. 3490–3496.

Acknowledgments

The support provided by Malaysian Ministry of Higher Education and Universiti Malaysia Pahang in the form of a research grant (RDU/UMP) vote number RDU1803160 for this study are highly appreciated.