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Physicochemical property enhancement of biodiesel synthesis from hybrid feedstocks of waste cooking vegetable oil and Beauty leaf oil through optimized alkaline-catalysed transesterification

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ABSTRACT

Recycling waste cooking vegetable oils by reclaiming and using these oils as biodiesel feedstocks is one of the promising solutions to address global energy demands. However, producing these biodiesels poses a significant challenge because of their poor physicochemical properties due the high free fatty acid content and impurities present in the feedstock, which will reduce the biodiesel yields. Hence, this study implemented the following strategy in order to address this issue: (1) 70 vol% of waste cooking vegetable oil blended with 30 vol% of *Calophyllum inophyllum* oil named as WC70CI30 used to alter its properties, (2) a three-stage process (degumming, esterification, and transesterification) was conducted which reduces the free fatty acid content and presence of impurities, and (3) the transesterification process parameters (methanol/oil ratio, reaction temperature, reaction time, and catalyst concentration) were optimized using response surface methodology in order to increase the biodiesel conversion yield. The results show that the WC70CI30 biodiesel has favourable physicochemical properties, good cold flow properties, and high oxidation stability (22.4 h), which fulfil the fuel specifications stated in the ASTM D6751 and EN 14214 standards. It found that the WC70CI30 biodiesel has great potential as a diesel substitute without the need for antioxidants and pour point depressants.

1. Introduction

The world population has reached 7.4 billion by the end of 2016 and it is forecasted that the world population will reach 12 billion by the end of the 21st century (Thompson, 2017). The rapid growth in world population leads to an ever-increasing demand for energy. Despite the escalating demands for energy, there is a bright future ahead for renewable energy, considering that global investments in renewable energy-based systems has increased by 5% compared with that in 2014, which amounts to USD 285.9 billion. This is more than twice the amount allocated for new coal and natural gas power-generation systems (USD 130 billion) (Arthouros, 2016). This indicates that there is an increasing demand for renewable energy-based systems compared with fossil fuel energy-based systems. In addition, low contract prices for renewable power purchase agreements has accelerated the use of renewable energy-based systems in developing countries (Aditiya et al., 2016a; Aditiya et al., 2016b; Hossain et al., 2017).

Biodiesels are a class of biofuels and as the name implies, biodiesels are alternatives to diesel for compression-ignition engines (Silitonga et al., 2013). Most of the first-generation biodiesels are derived from edible feedstocks such as palm, canola, and corn oils. Normally first-generation biodiesels possess poor cold properties and good oxidation stability due to high level of saturated fatty acids (Sierra-Cantor and Guerrero-Fajardo, 2017). But, the sustainability of these biodiesels is a major issue because these fuels lead to competition between the use of edible vegetable oils for food and fuel production (Milano et al., 2016). Therefore, much effort is being made to produce biodiesels from non-edible vegetable oils such as *Jatropha curcas* (Takase et al., 2015; Wang et al., 2011), *Madhuca indica* (Saravanan et al., 2010), *Calophyllum inophyllum* (Azad et al., 2016; Damanik et al., 2017), *Ceiba pentandra* (Kusumo et al., 2017; Putri et al., 2012; Yunus Khan et al., 2015), *Sterculia foetida*, *Sapium sebiferum* (Wang et al., 2011), *Euphorbia lathyris* (Wang

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Nomenclature

CFPP	cold filter plugging point	SF	*Sterculia foetida* oil
CI	*Calophyllum inophyllum* oil	WC	waste cooking vegetable oil
CP	*Ceiba pentandra* oil	WC70CI30	70 vol% of waste cooking vegetable oil + 30 vol% of *Calophyllum inophyllum* oil
CP	cloud point	WC70CP30	70 vol% of waste cooking vegetable oil + 30 vol% of *Ceiba pentandra* oil
FAME	fatty acid methyl ester	WC70JC30	70 vol% of waste cooking vegetable oil + 30 vol% of *Jatropha curcas* oil
FFA	free fatty acid	WC70SF30	70 vol% of waste cooking vegetable oil + 30 vol% of *Sterculia foetida* oil
FTIR	Fourier transform infrared		
GC	gas chromatograph		
JC	*Jatropha curcas* oil		
PP	pour point		

et al., 2011; Zapata et al., 2012), *Reutealis trisperma* (Riayatsyah et al., 2017), and *Pongamia pinnata* oils (Sharma et al., 2009). These biodiesels are known as second-generation biodiesels and they have been proven to be potential diesel substitutes (Atabani and César, 2014; Silitonga et al., 2018). The findings are indeed promising because the physicochemical properties of these biodiesels are comparable to those for diesel (Nanthagopal et al., 2016; Silitonga et al., 2014). Second-generation biodiesels are also favourable because of their good cold flow properties (*Jatropha curcas* ~ CFPP (2 °C) (Sarin et al., 2007), *Madhuca indica* ~ PP (3 °C) (Muthukumaran et al., 2017), *Calophyllum inophyllum* ~ CP (0 °C) and PP (0.8 °C) (Silitonga et al., 2016), *Nigella sativa* ~ CP (−1), PP (−1) and CFPP (−4) (Yunus Khan et al., 2015) and higher oxidation stability (*Calophyllum inophyllum* ~ 15.17 h (Silitonga et al., 2016), *Jatropha curcas* ~ 9.41 h, (Silitonga et al., 2013)), as well as lower kinematic viscosity and acid value (*Ceiba pentandra* ~ 0.16 mg KOH/g and *Nigella sativa* ~ 0.26 mg KOH/g (Yunus Khan et al., 2015)) (Islam et al., 2016). Fatty acid methyl ester is one of the method to determine the cold flow properties, oxidation stability or storage stability. Generally, high oxidation stability is dependent on the saturated fatty acids, but high saturated fatty acids will lead to poor cold filter plugging point. This is because the cold properties are more dependent on the unsaturated fatty acids of the feedstocks (Ramos et al., 2009). However, FAME content could not solely determine the cold properties and oxidation stability of the non-edible oil. Silitonga et al. found that *Jatropha curcas* possess good cold flow properties and also good oxidation stability. Moreover, other researchers found that there are other factors that will affect the oxidation stability of the biodiesel. These factors are the configuration of the double bond, chain length, branching of the chain, molecular weight, presence of impurities such as metal, free fatty acids, additives, antioxidants, and water contamination (Pullen and Saeed, 2012). To acquire best cold properties and oxidation stability, mixing two feedstocks can be performed in order to have a balance of saturated and unsaturated fatty acid methyl ester.

However, the cultivation of non-edible plants is not economically feasible in the long term unless the non-edible plants are cultivated on non-arable or marginal lands. Hence, there is a critical need to explore alternative biodiesel feedstocks that are economical and sustainable in the long term, and this has been the focus of many studies in recent years. Undoubtedly, waste cooking (WC) vegetable oils are one of the biodiesel feedstocks that fulfil these criteria (Diaz and Borges, 2012; Karmee et al., 2015). WC oils are typically disposed after use on a daily basis and the disposal of millions of tonnes of WC oils is a critical issue in the food and beverage industry around the world (Sabudak and Yildiz, 2010). Recycling WC oils is a feasible solution to address this issue since this will prevent the oils from being disposed into the landfills. Recycling WC oils by reclaiming and using these oils as biodiesel feedstocks is one of the ways to achieve this goal.

Much effort has been made to convert WC oils into biodiesels (Diaz and Borges, 2012; Mahesh et al., 2015; Maneerung et al., 2016; Sirisomboonchai et al., 2015; Yahya et al., 2016). However, using WC oils as biodiesel feedstocks poses a significant challenge because of their free fatty acid (FFA) content as well as the presence of water and impurities, all of which will lead to low biodiesel yields due to saponification during transesterification. Various catalysts have been used in transesterification of WC oils such as homogeneous catalysts (Demirbas, 2009; Felizardo et al., 2006; Lam et al., 2010; Sabudak and Yildiz, 2010), heterogeneous catalysts (Corro et al., 2016; Gupta et al., 2015; Tan et al., 2015a; Tan et al., 2015b), enzymatic catalysts (Charpe and Rathod, 2011; Mehrasbi et al., 2017) and other types of catalysts (Caldas et al., 2016; Liang et al., 2013; Ullah et al., 2015). Even though most WC biodiesels fulfil the fuel specifications stipulated in the ASTM D6751 and EN 14214 standards, the main issues of WC biodiesels are low biodiesel yields (Yahya et al., 2016), poor oxidation stability (Fu et al., 2016; Qu et al., 2016; Savi et al., 2017), high kinematic viscosity, and poor cold flow properties (Xue et al., 2016).

One of the ways that can be used to improve the physicochemical properties and cold flow properties of WC biodiesels is to blend WC oils with other types of feedstocks (Canoira et al., 2008; Dharma et al., 2016; Yunus Khan et al., 2015). In this study, the following strategy was implemented in order to address the main issues associated with WC biodiesels. Firstly, WC oil was blended with other non-edible vegetable oils (*Calophyllum inophyllum* (CI), *Jatropha curcas* (JC), *Ceiba pentandra* (CP), and *Sterculia foetida* (SF) oils) at a ratio of 7:3. The most suitable crude oil blend was selected for biodiesel production based on the physicochemical properties. Secondly, a three-stage process was implemented that consist of two pre-treatment processes and transesterification. There process are needed to reduce the FFA content, water and impurities of the crude oil mixure. Thirdly, the transesterification process parameters (methanol/oil ratio, reaction temperature, reaction time, and catalyst concentration) were optimized in order to obtain high biodiesel yield, and examine the effect of the studied parameters towards the conversion yield.

Next, a quadratic model was developed to precisely predict the biodiesel conversion yield as function of the aforementioned transesterification process and the reliability of the prediction model by analysis of variance (ANOVA) was verified. The biodiesel was characterized using Fourier-transform infrared (FTIR) spectroscopy in order to identify the functional groups present in the biodiesel, which will provide insight on the production of methyl esters from the triglycerides. Finally, the physicochemical properties of the biodiesel produced in this study was measured and compare them with specifications stated in the ASTM D6751 and EN 14214 standards.