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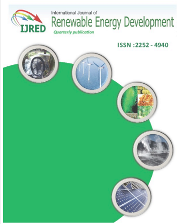

Research Article

Corrosion of The Metal Parts of Diesel Engines In Biodiesel-Based Fuels

Xuan Phuong Nguyen* and Hai Nam Vu

Ho Chi Minh city University of Transport, Ho Chi Minh City, Vietnam

ABSTRACT. Biodiesel, an environmentally-friendly bio-fuel, has been regarded as one of the most promising alternatives to fossil fuels whose use is rampant in the transportation sector. However, it is important that the corrosive effects of this fuel on engines are studied. This work reviews the corrosiveness that biodiesel exerts on various engine components, especially those made out of metals. First, an analysis of the corrosion mechanisms of metals exposed to biodiesel is provided. The conventional and advanced analysis methods will be applied to measure the level of corrosiveness in static immersion test, and to assess the formation of secondary products, if any, in biodiesel and any metal strips in contact with biodiesel-based fuel. The use of inhibitions to guard against corrosion will be mentioned. Lastly, several significant causes of metal corrosion, namely, the presence of dissolved oxygen and oxidation products, TAN change, a rise in dissolved water, the presence of metals, and the changes in biodiesel properties will also be presented.

Keywords: biodiesel, corrosion, metal, analysis techniques, inhibitor

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1. Introduction

The rapid depletion of fossil fuels has prompted a global search for new energy sources. Various sources have been studied. Of those, biofuels, which are mostly made up of biogasoline and biodiesel, are seen as the most feasible and having the potential to rival fossil fuels. Worldwide, 64 countries have approved the use of biodiesel as an energy source for vehicles. Air pollution, a global phenomenon, is caused mainly by exhaust emissions from internal combustion engines (ICE) that use fossil fuels (Hoang *et al.* 2017). Also, as energy demand is projected to go up by 40% by 2030, roughly 16.8 tons each year(Xia 2016; Beyene *et al.* 2018), the supplies for fossil fuels will be tremendously strained. In order to minimise the adverse effects of fossil fuel shortages, two solutions related to ICE have been proposed with the first being the engine design and the second being the quest for alternative fuels (Demirbas 2017; Hoang 2018a). Both are ways to improve the energy efficiency of engines and to reduce the amount of toxic emissions(Hoang 2018b; Hoang 2019). Bio-based fuels, with their renewability and other advantages, have been considered a promising candidate. They can be divided by origin, the first being vegetable-oil-based, the second being animal-fat-based and biomass-based(Hoang et al. 2019a;Hoang et al. 2019b). Among bio-based fuel, several researchers about the biodiesel production feedstock and use of biodiesel for diesel engine have been studying. Obviously, biodiesels have been presenting many comparative advantages to fossil fuels (Hoang et al. 2019c; Takase et al. 2018). Despite major comparative advantages to fossil fuels, biodiesel itself has many inherent drawbacks, such as corrosion, tribo-corrosion phenomenon, instability and other environmental factors(Al-Dawody et al. 2013;Pantoja et al. 2013;Fazal et al. 2014;Hoang et al. 2019).

According to Nernst, all metal elements tend to move into solutions. However, the mass of metal matters and the extent to which corrosion happens differ for wear metals and metal ions. These two factors are dependent on the abrasion, the oxidation potential, the properties of a fuel. In general, the presence of metals in the fuel will lead to corrosion(Singh et al. 2012). Due to these reasons, the biggest obstacle that the manufacturers must face is the failure of the mechanical parts of engines running on biodiesel. These reasons explain why the biggest obstacle that manufacturers face is the failure of mechanical parts of engines running on biodiesel. These parts are often the static components of the fuel system such as fuel take, filter, supply pumps, injector, fuel line, exhaust system, cylinder liner, etc. Some moving components are also affected, too, such as piston crown, piston rings, valve, plunger, connecting rod…etc(Haseeb *et al.* 2011;Hoang *et al.* 2019e). The components of diesel engines, the fuel system, and commonly-used materials for fabrication are shown in Fig.1

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* Corresponding author: phuong@ut.edu.vn

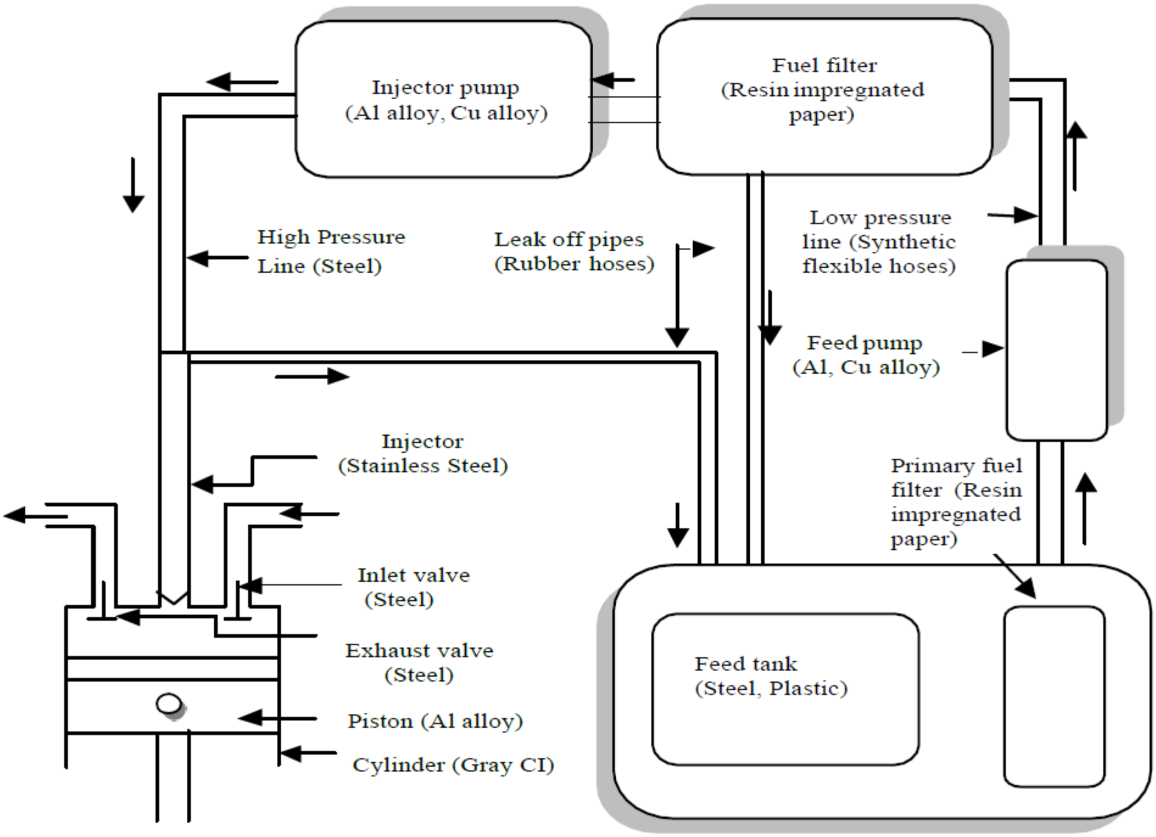


Fig. 1. Used material for fabrication of parts in diesel engines (Sorate et al. 2013)

Recently, the corrosiveness of biodiesel on diesel engines has been assessed to determine the viability of the use of biodiesel for diesel engines. It turns out that without sulphur content, the corrosion levels in biodiesel-based-fuel containers will be reduced(Qiu et al. 2011;Hoang et al. 2018c). During the production process, acid or alkali catalysts in the homogeneous or heterogeneous states are often used to boost the esterification efficiency (Aysu et al 2016). One of those catalysts is sulphuric acid. However, the acid itself will cause corrosion in biodiesel-powered engines(Su et al. 2014). In order to overcome this challenge, solid acid catalysts are suggested because they can be separated much more easily from biodiesel after esterification process(Busca 2010;Sharma et al. 2011). Furthermore, a high level of purity is required in biodiesel because it improves the compatibility between the engine and the fuel. Therefore, impurities from the esterification process such as glycerol, fatty acids, alcohol, and catalysts may be the origins of deposit formation, corrosion and fuel failure (Nagy et al. 2009; Hoang et al. 2019f). On the other hand, because biodiesel is much more lubricating than fossil fuels, the former is more likely to witness the dissolving of the metallic matters of mechanical parts. This necessitates a search for corrosion inhibitors to enhance the endurance of biodiesel-powered engines (Singh et al. 2012). An elaborate review of Haseeb et al. (Haseeb et al. 2011) and a careful evaluation of Sorate et al (Sorate et al. 2013) on the effects of biodiesel on the materials durability in diesel engine components and fuel system (Fig.1) fabricated by many material types based on metals or alloys (such as steel, stainless steel, copper/copper-based alloy, aluminum/aluminum-based alloy, cast iron..etc), and non-metallic materials (such as elastomer, plastics, rubber, ceramic fiber...etc) have shown a strong correlation between the found impurities in biodiesel, biodiesel deterioration and the corrosiveness through oxidation (Jakeria et al. 2014). Thus, the corrosion of materials in the engines fuelled by biodiesel needs to thoroughly researched.

2. Composition of biodiesel

Esterification reactions between triglycerides (vegetable oils/animal fats) and alkanol (methanol or ethanol) lead to the formation of biodiesel. Therefore, biodiesel is, in essence, an oxygenated mono-alkyl ester with at least two oxygen atoms. However, the efficiency of esterification reactions can never be as high as 100%, and different catalysts such as homogeneous alkaline types or acid catalysts have been used to enhance both the reaction efficiency and the yields. Table 1 lists the reaction efficiency of esterification and as-used catalyst types along with as-produced biodiesel types based on standard EN 14214 which requires the around 96.5% of ester minimum amount in biodiesel ("Biodiesel Standards, EN 14214:2003," n.d.).

Table 1 shows that a certain amount of impurities exists in biodiesel. They are often unused catalysts, monoglyceride, diglyceride or triglyceride, free fatty acid, water, etc. The residue, especially those formed by acid catalysts can corrode various materials aggressively. As can be seen from Table 1, alkaline catalysts have higher ester reaction efficiency that acid ones.

Table 1.
As-used catalysts and yields of biodiesel production from various feedstocks

References	Catalyst types	Feedstocks	Yields (%)
Felizardo et al. 2006	NaOH	Waste oil	98
Ji et al. 2006		Soybean oil	100
Lubes et al. 2009		Palm oil	95
Dias et al. 2008		Sunflower oil	97.1
Tupufia et al. 2013		Coconut oil	91
Zagonel et al. 2002	KOH	Soybean oil	95
Elsheikh et al. 2011		Palm oil	98.4
Moradi et al. 2013		Soybean oil	93.2
Yihuai et al. 2011		Soybean oil	92.4
Ghadge et al. 2005	H_2SO_4/ KOH	Mahua oil	98
Patil et al. 2009		Jatropha curcas oil	90-95
Kumartiwari et al. 2009		Jatropha curcas oil	99
Tashtoush et al. 2004	H_2SO_4	Animal fat	75
Kouzu et al. 2008	CaO	Waste oil	99
Wuttichai et al. 2006		Palm oil	97
Yie et al. 2015	Ostrich-eggshell-based-CaO	Cooking oil	94-96
Samart et al. 2010	CaO/silica	Soybean oil	95.2
Chew et al. 2008	CaO/SBA-14	Sunflower oil	95
Rashid et al. 2009	$NaOCH_3$	Cotton oil	96.5
Rashid et al. 2016		Cannabis sativa oil	86
Shin et al. 2012	Non-catalyst	Rapeseed oil	93.6
Thirugnanasambandham et al. 2017	Microwave	Waste frying oil	97
Osama et al. 2017	Ultrasonic	Eucalyptus oil	95.7

Although biodiesel and fossil diesel fuel have some common properties, they also differ significantly in terms of molecular structure, oxygenated moieties, molecular components, stability, physical properties, and especially total acid number (TAN). It is also worth pointing out that high levels of TAN are considered the primary cause of degradation of the exposed metal surface (Fazal et al. 2014). Also, because alkaline catalysts may reduce TAN of biodiesel, affinities for metal surfaces, and as a result, corrosion, alkaline catalysts are logical and suitable options for biodiesel production. The results of the study given in Table 1 show the strong effects of the choice of catalyst type on the yield. To be more specific, NaOH catalyst is the waste oil, soybean oil, J. curcas oil and sunflower oil, but not for palm oil, coconut oil because yield is lower than the requirement of EN 14214 standard. To conclude, catalyst types, feedstock, and production process are the three primary factors that contribute to the composition of biodiesel.

3. Mechanism of corrosion of metals in biodiesel

At present, reports that explain and prove that corrosion mechanism of metals in biodiesel are scarce. As for the corrosion mechanism for copper, Fazal et al.(Fazal et al. 2010) reported the pitting corrosion phenomenon of copper in biodiesel at 80°C. The relative concentrations of two ion types, being positive ions and negative ions, strongly affected the pit morphology and the mechanism of the bites on the copper surface. The oxygen presence in biodiesel resulted in the formation of $CuO/CuCO_3$ on the outer layer, and Cu_2O on the inner layer (Zuleta et al. 2012). The pits appearing on the copper surface were thought to be formed by the substitution of oxygen ions in Cu_2O composition to CuO because of the low level of instability of Cu_2O (Hernández et al. 2010). The presence of other substances and ion groups such as dissolved water, CO_2, RCOO-, etc… was considered the main cause of the formation of the carbonate and hydroxyl based copper compounds ($CuCO_3$, $Cu(OH)_2.CuCO_3$, $Cu(OH)_2$). The study of Fazal et al.(Fazal et al. 2013) has indicated that the dissolved components such as O_2, H_2O, CO_2,and $RCOO^-$ in biodiesel had an association with formed copper compounds. CO_2 and $RCOO^-$ components were supposed to be the origination of carbonate formation after immersion time. The mechanism corrosion of metals was presented in reactions from 1-8 with complex $M_xO_yH_zC_m$, M^{x+}, $M_2(CO_3)_{2y/x}$, $M(OH)_m$ being considered the corrosion products (Zuleta et al. 2012; Fazal et al. 2013).

$$\text{Air: } O_2 + 2H_2O + 4e^- \rightarrow 4OH^- \quad (1)$$

$$2H_2O + 2e^- \rightarrow H_2 + 2OH^- \quad (2)$$

$$\text{Acid: } RCOOH \rightarrow RCOO^- + H^+ \quad (3)$$

$$\text{Metals: } 2xM + yO_2 \rightarrow 2M_xO_y \quad (4)$$

$$M_xO_y + 2yH^+ \rightarrow xM^{x+} + yH_2O \quad (5)$$

$$2M_xO_y + yCO_2 \rightarrow xM_2(CO_3)_{2y/x} \quad (6)$$

$$4M + mO_2 + 2mH_2O \rightarrow 4M(OH)_m \quad (7)$$

$$M_xO_y + 4M(OH)_m + xM_2(CO_3)_{2y/x} + H^+ + O_2 + CO_2 + H_2O \rightarrow M_xO_yH_zC_m \quad (8)$$

According to Gil et al (Gil et al. 2007), the presence of a thin aqueous layer (1 µg/cm²) was the prerequisite to the formation of the hydroxyl bonds for copper oxide. Nevertheless, some published studies confirm the need for further investigation into the copper corrosion in biodiesel. In order to assess the corrosion levels of steel in biodiesel, X-ray diffractometry (XRD) method was used because iron is the main component of the material. An analysis of XRD detected the presence of $Fe(OH)_3$, $Fe_2O_2CO_3$ and Fe_2O_3 compounds. These compounds were formed along with oxygen and water presence. $Fe_2O_2CO_3$ was the chemical reaction product between H_2CO_3 and FeO(OH), whereas FeO(OH) was the product produced by the redox reaction between Fe, O_2 and H_2O (Fazal et al. 2011c). The absorption of CO_2, O_2, and H_2O from air into biodiesel was thought to cause the formation of some corrosive factors like H_2CO_3. The corrosion of carbon steel, also, may be originated from the oxygenation of biodiesel. Fatty acid components in biodiesel, themselves, have led to oxidised biodiesel, while the absorbed water increased along with the increase in temperature (Haseeb et al. 2010).

Alkaline components such as KOH and NaOH, in biodiesel as catalysts are considered themain cause of the corrosion aluminium or aluminium-based alloys. When in contact with alkaline catalysts, AlO- and $Al(OH)^{-4}$ ions are released to form a passive layer of $Al(OH)_3$ via the electrolyte reduction such as water, or methanol, resulting in hydrogen evolution (Hu et al. 2012). The morphological corrosion of metals is shown in Fig.2.

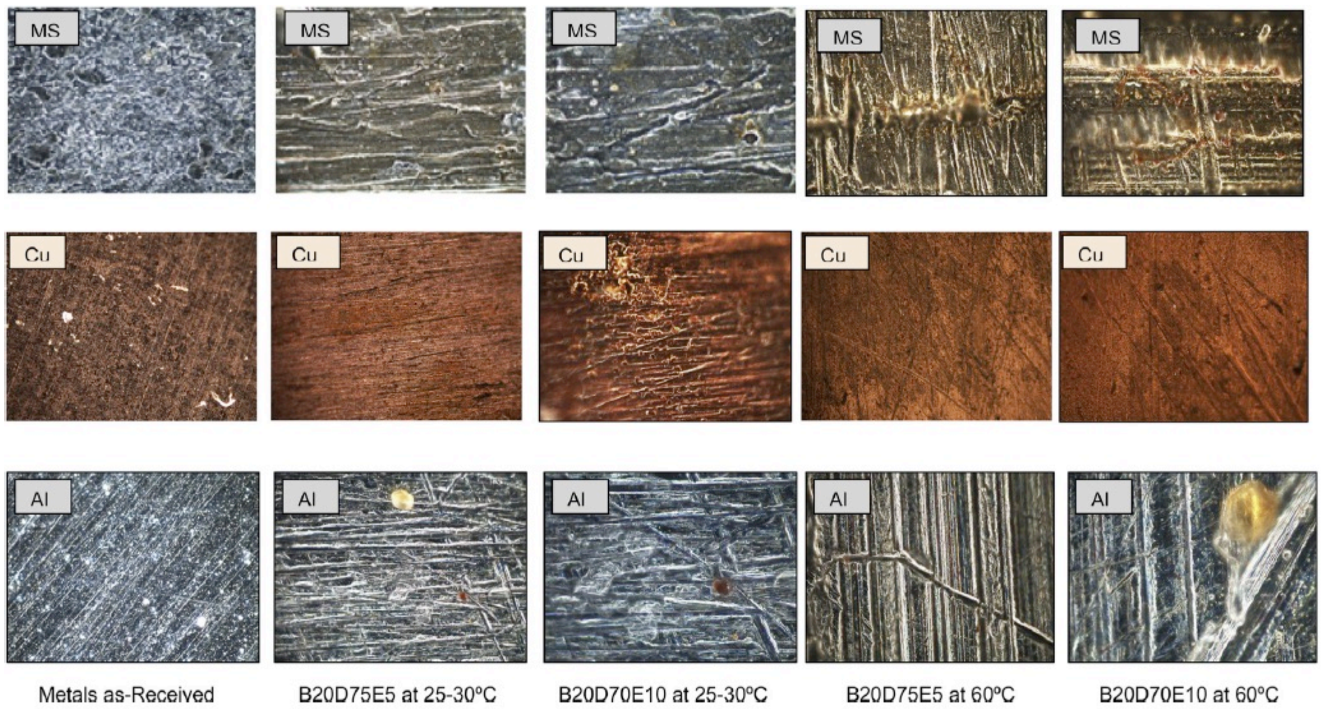


Fig. 2. The morphological corrosion of copper, aluminium, and mild steel after exposure to biodiesel-diesel-ethanol blends at 60 °C (Saravana et al. 2016)

In general, some leading causes of metal corrosion in biodiesel may be included: the presence of dissolved oxygen and oxidation products, TAN change, increase in dissolved water, the metal presence, the changes of biodiesel properties.

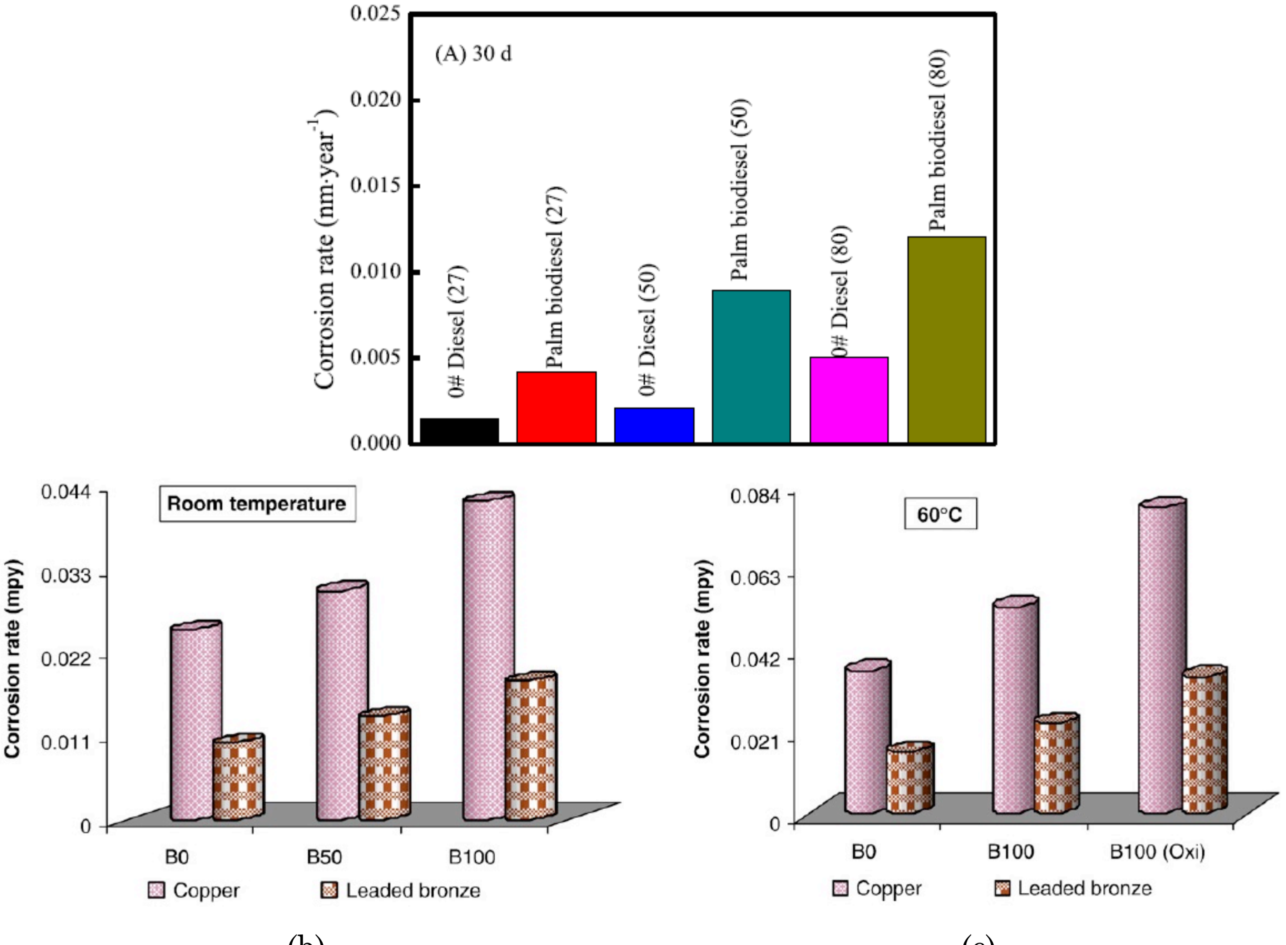
4. Effect of biodiesel on the corrosion behavior of metallic materials

Biodiesel use for diesel engines has been a relatively successful story due to the increased engine performance and emissions characteristics. However, the corrosiveness of exposed metals and degradation of biodiesel continue to baffle researchers worldwide because it must take a long time to test the matters related to corrosiveness, degradation, the durability of mechanical parts and biodiesel-based fuel. Nowadays, many parts of diesel engines and fuel system require high precision and corrosion resistance. Corrosion damage of parts in the fuel supply system dramatically increases with oxidised biodiesel and moisture-absorbed biodiesel. Several studies have been conducted on the ease of water absorption and microorganism contamination of biodiesel in comparison with fossil diesel(Díaz-Ballote et al. 2009; Kamiński et al. 2008). These allow the electrochemical corrosion processes to be understood more easily. Currently, to measure the corrosion level of biodiesel, ASTM D130 is used. In this method, a copper strip is immersed in biodiesel during a specific temperature and time. After that, it is removed and washed to evaluate by observing the colour of the copper strip(Geller et al. 2010). Another standard, being ASTM D93, is also used to test the copper strip corrosion(Singh et al. 2012). Nonetheless, both methods as per ASTM D130 and ASTM D93, as well as the method of titration to determine the TAN value, are not reliable in evaluating the corrosion level of individual organic acids. In general, corrosion level depends on unsaturated molecules, FFA, hygroscopic nature of biodiesel and types of materials contacting with biodiesel. The copper-based alloys are more corrosive than the iron-based alloys (Fazal et al. 2011a). As presented in Fig.1, most engine parts and fuel system components are made from metals and alloys based on aluminium, copper, iron and stainless steel, and these materials are prone to corrosion. In small diesel engines, aluminium/aluminium-based alloys can be used for some components such as piston components, cylinder heads, engine blocks; copper and its alloys are used for fuel pump components and injector components; nozzle, fuel filter, valve bodies, and pump rings are composed of stainless steel (Fazal et al. 2010; Pham et al. 2017). To evaluate and test the corrosiveness of as-mentioned metals/alloys, the observation of biodiesel colour is probably the simplest method. Other methods include the static immersion test (SIT), the immersion in various temperature conditions may also be considered. Also, the assistance of novel and modern equipment for taking or analysing the photographs of microstructure in the surface which aim at obtaining the exact result are using widely. Sgroi et al. (2005) reported the use of biodiesel for a diesel engine with a focus on injector and burner filter components. High chrome stainless steel was used to fabricate the injector; meanwhile, copper-based alloys and copper were used for burner filter components. As a result, the copper content found in biodiesel fuel after 2 hours of the test increased from 0.1 to 21 ppm. The corrosion factors also were observed on the bronze filter, even including the pitting corrosion, after 10 hours of using biodiesel that was preheated to 70°C. However, it did not detect any corrosion factors on the injector.

Fazal et al. (2011c) examined the corrosion properties of mild steel after immersing the material into different fuels, including B0 (100% diesel fuel), B50 (50% palm biodiesel and 50% diesel fuel), B100 (100% palm biodiesel), in the SIT with various temperatures, being room temperature, 50°C and 80°C during 1200h. The method of weight loss measurement and the observation of surface changes were used to evaluate the corrosion level. After testing time, the obtained results showed that the water content and oxidation products in biodiesel increased proportionally to temperature as mild steel came into contact with biodiesel resulting in the corrosion level also being increased along with the increase in temperature (Fig.3). Fazal et al. (2010) explored the effects of palm oil-based biodiesel on the corrosiveness behaviours of various materials in the automobile. The test conditions composed of the metal/alloy samples such as copper (99.99% of purity), aluminium (99% of commercial purity), 316 stainless steel (18% of chromium, 11% of nickel, 2% of manganese, 1% of silica and 0.08% of carbon), along with fuels including B0 (0% of palm biodiesel) and B100 (100% of palm biodiesel). Metal/alloy samples were immersed in B100 and B0 at temperature 80°C and stirred at the speed of 250 rpm by a magnetic stirrer. A conclusion on the corrosion to copper and aluminiumwas reported; however, stainless steel seemed not subject to corrosion. To be more specific, after 1200 hours of test time, the corrosion rate of the sample as being immersed in B100 for copper was 0.586mpy, and for aluminium only 0.202 mpy, and for stainless steel only 0.015mpy. These corrosion rates for B0 were 0.3 mpy for copper, 0.15mpy for aluminium, 0.015 mpy for carbon steel. Moreover, some biodiesel properties such as density and viscosity seemed to remain relatively unchanged after exposure to metals (Fig.4). Hence, aggressive corrosion along with a biodiesel degradation has been shown(Fazal et al. 2010;Cao et al. 2007).

Hu et al. (2012)have conducted an experiment to evaluate the corrosion properties of metals, including copper, mild carbon steel, stainless steel, andaluminium, in biodiesel compared with fossil diesel fuel. After being immersed in biodiesel and fossil diesel fuel for 60 hours at 43°C, the corrosion rates of the used metals in biodiesel were higher than in fossil diesel fuel (Fig.2). In addition, the copper and mild carbon steel corrosion rates were also significantly higher than those of aluminium and stainless steel. The similarity of copper corrosion properties was also agreed by Geller et al.(Geller et al. 2010), and Aquino et al (Aquino et al. 2012) who have indicated that copper and copper-based alloys (brass) were prone to corrosion after carrying out an experiment related to weight loss due to pitting corrosion. Haseeb et al.(Haseeb et al. 2010) tested the effects of palm biodiesel on the corrosion level of copper and leaded bronze in two different conditions. The first test was conducted at room temperature for B0, B50 and B100 for 2640 hours. The other was carried out with B0, B100 and B100 (oxidised) at 60°C for 840 hours. From the results of the first test condition, the rate of corrosion level of copper for B100 was determined to be 0.042mpy (milligram per year), of bronze for B100 was 0.018mpy. With the second test condition, this rate of copper corrosion for oxidized-B100 was 0.053mpy, of bronze corrosion for oxidized-B100 was 0.023mpy. In addition, this study's authors also thought that the tin (Sn) presence in bronze might be an agent enhancing the corrosion resistance level compared to copper. The similar results about corrosion rates of metal in biodiesel were also reported. To be more specific, the corrosion rate of copper (0.323615mpy) was higher than that of mild carbon steel (0.170124mpy) and aluminium alloys (0.162201mpy) at room temperature. This corrosion rate also increased along with the increase in biodiesel concentrations in a blend with diesel fuel (Cursaru et al. 2014). In a study of Jin et al.(Jin et al. 2015), ASTM 1045 mild steel was used to immerse statically in pure palm biodiesel and diesel fuel at various temperatures, being 27°C, 50°C, and 80°C, in order to assess the effect of fuel on the corrosiveness rate. After 30, 60 and 120 days of the test, the sample was removed from fuel. The results have demonstrated that palm oil-based biodiesel was more corrosive than fossil diesel fuel.

Another common method to measure corrosion levels is weight loss. In another study conducted by Kaul et al.(Kaul et al. 2007), the effects of different feedstock-based biodiesels and at different temperatures (from 15 to 40°C) on the corrosion behaviour of the diesel engine piston fabricated by aluminium alloys were tested in SIT. The study showed that the corrosion phenomenon for piston liner and piston metal occurred after being immersed in biodiesel (including jatropha curcus (S1)-, kanarja (S2)-, mahua (S3)-, salvadora-based biodiesel (S4)). The weight loss of piston liner caused by corrosiveness for S1 was 3.6mg, for S2 was 0.3mg, for S3 was 0.3mg and for S4 was 6.1mg. The corresponding corrosion rate for S1 was 0.0117mpy, for S2 was 0.0058mpy, for S3 was 0.0058mpy and for S4 was 0.0136mpy in comparison with 0.0058mpy of diesel fuel. A high weight loss of piston metal for S4 could be seen, at 2.1 mg compared to 0.2mg for S1 and 0.1mg for S2, S3. The above results could be explained by high TANs of S1 and S4, which increased corrosion.



(b) (c)

Fig. 3. The relationship between temperature and corrosion rates, a-from Reference (Jin et al. 2015); b- at room temperature, c- at 60°C (Haseeb et al. 2010)

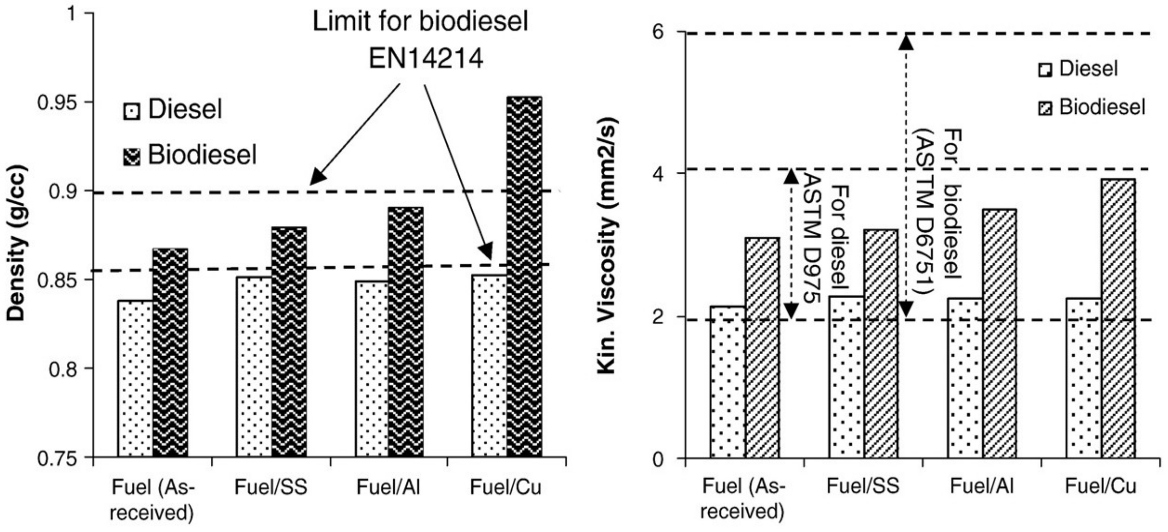


Fig. 4. Changes in fuel properties (diesel fuel and biodiesel) before and after 1200h exposing to metals at 80°C (Fazal et al. 2010)

The optical photographs of tested metallic surfaces such as copper, aluminium, and stainless steel could be found in the reference (Fazal et al. 2010). Similarly, Sorateet al.(Sorate et al. 2013) have presented the results of weight loss of metals from parts such as top ring, compression ring, scrap ring and oil ring after exposing them to biodiesel in comparison with fossil diesel fuel. To sum up, the measurement methods based on weight loss and corrosion rate have proven and shown that the corrosion properties of metals in biodiesel were higher than those of fossil diesel fuel (Fig.5). Moreover, the colour of biodiesel after exposing to metals was used to evaluate the corrosion level. This method was also found in references (Fazal et al. 2013)(Haseeb et al. 2010)(Hu et al. 2012)(Fazal et al. 2011b).

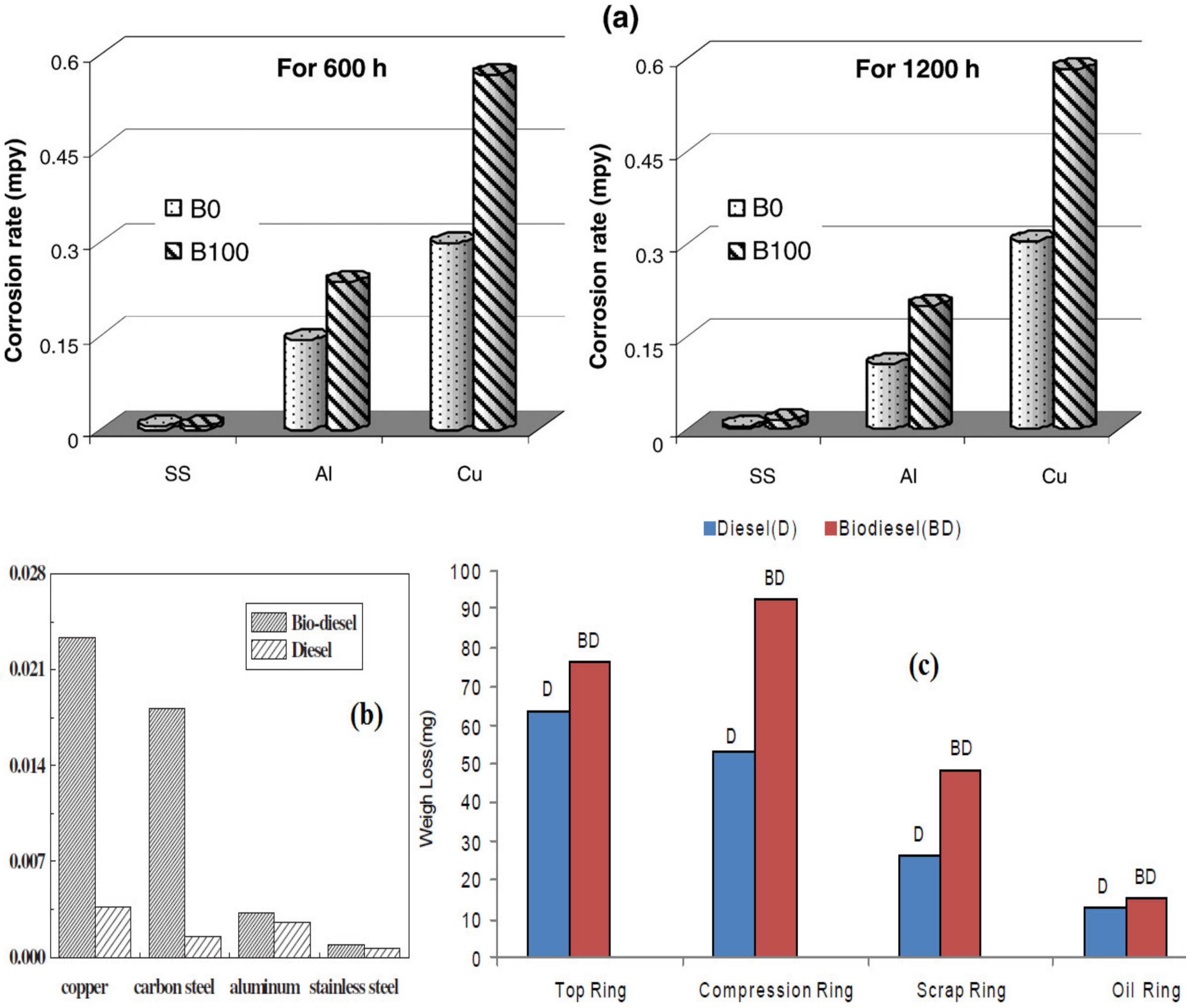


Fig. 5. Corrosion property of metals in biodiesel and fossil diesel fuel based on measurement methods of corrosion rate and weight loss, (a)-Reference (Fazal et al. 2010),(b)- Reference (Hu et al. 2012), (c)-Reference (Sorate et al. 2013)

Recent studies have also adopted advanced technologies based on SEM/EDS, AAS, XPS, EIS, FT-IR, Raman spectra analysis. The use of electrochemical impedance spectroscopy (EIS) to observe the corrosion properties of metals has proven that steel was more resistant to corrosion compared to copper and its alloys because the former has carbon content within the range of 0.2 to 2.1% by mass along with high corrosion resistance of carbon. Also, stainless steel with types of single-phase or double-phases was considered the alloys with high resistance to corrosion because of the absence of the differential of electrode potential (Pham et al. 2017; Cao et al. 2007). Nonetheless, a report of Prieto et al. (2008) has indicated that galvanic metal corrosion in steel could occur because the conductive electric of biodiesel was found higher than that of fossil diesel fuel. Maru et al. (2009) conducted an experiment in which strips of structural carbon steel (CS) were immersed into three types of fuel, namely, soybean oil-based biodiesel-, sunflower oil-based biodiesel, and diesel fuel in static emersion conditions for 115 days. After the first 60 days, the results from strips in biodiesels and diesel fuel showed that the materials were more compatible to soybean oil-based biodiesel than to sunflower oil-based biodiesel and diesel fuel because the CS strips weight did not change. After 115 days, the weight loss of test metal samples in diesel fuel and biodiesel were quite low (around 10^{-5}g). However, Fourier Transform Infra Red (FT-IR) and Raman spectra analysis (Fig.6) have shown the formation of secondary products originated from fuel degradation.

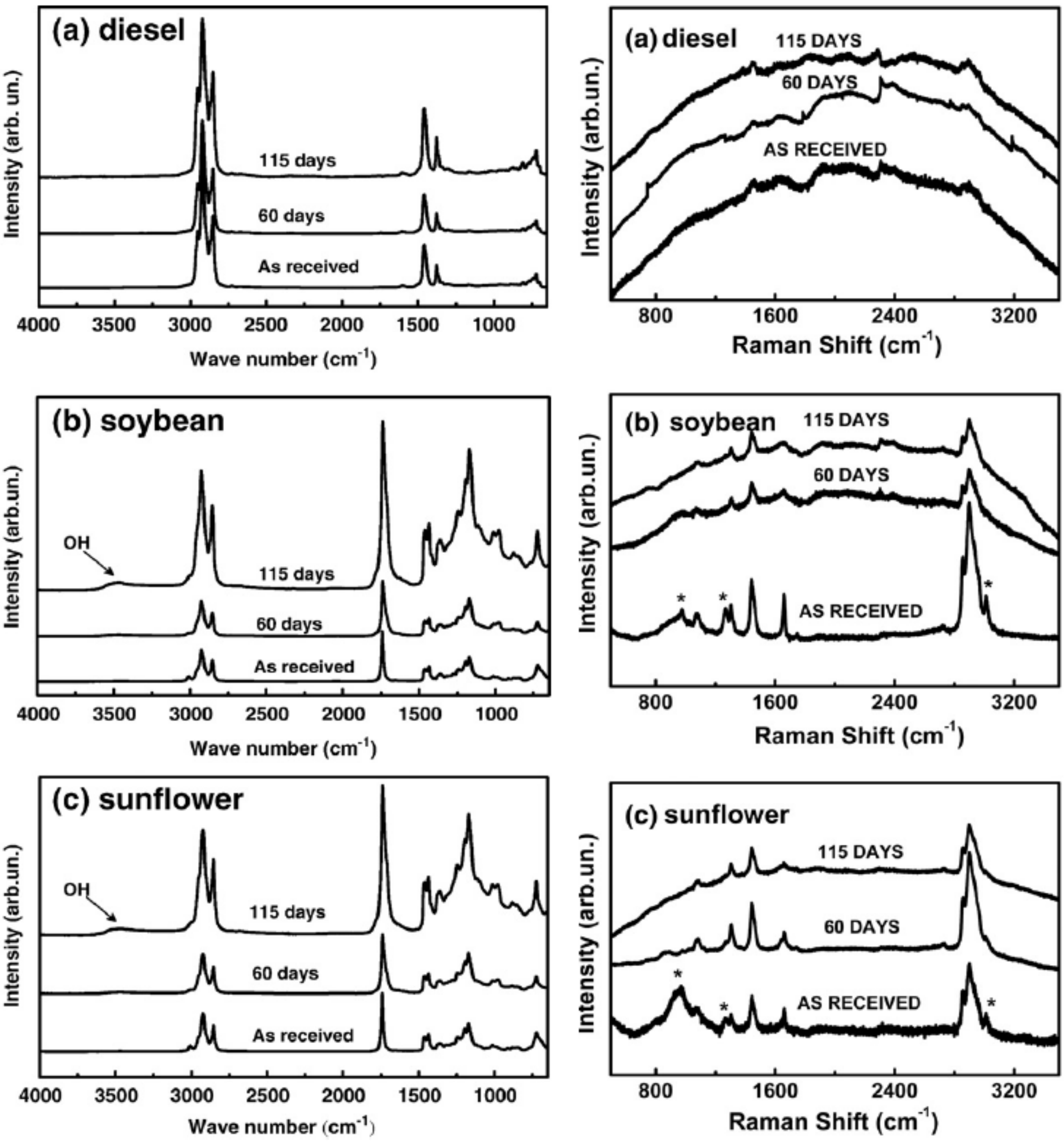


Fig. 6. FT-IR and Raman spectra of fuel before and after immersion tests(a) Diesel; (b) Soybean; (c) Sunflower (Maru et al. 2009)

The broadened C=O peak at 1740 cm^{-1} thanks to the mutual overlapping of aldehyde-, and ester carbonyl groups (FT-IR spectra), and a symmetric plane bending modes of unconjugated (=C-H) bonds (Raman spectra) with the peak at 3313 $cm^{-1}$was reported. This proved that soybean oil-based biodiesel caused more ageing than sunflower oil-based biodiesel. These results were the same as those from a study of Hu et al. (2012). In another study, ASTM 1045 mild steel was immersed in palm oil-based biodiesel (Jin et al. 2015). The analysis of the results based on FT-IR spectral performed after 120 days showed the main corrosion products on used samples from biodiesel, namely α-FeOOH (1372.27 cm^{-1}), β-FeOOH (881.84 cm^{-1}), δ-FeOOH (465.62 cm^{-1}, 1196.93 cm^{-1}, 1436.81 cm^{-1}), and $Fe_2O_2CO_3$ (609.17 cm^{-1}). While the products of corrosion process from fossil diesel were primarily α-FeOOH (1372.81 cm^{-1}), γ-FeOOH (1022.00 cm^{-1}), δ-FeOOH (11196.81 cm^{-1}) and $Fe_2O_2CO_3$ (609.23 cm^{-1}), these results have proven the presence of compounds such as FeO, Fe_2O_3, FeO(OH), $FeCO_3$, $Fe_2O_2CO_3$on the surface of the biodiesel-immersed sample. However, the absence of $FeCO_3$, $Fe_2O_2CO_3$compounds could be detected on the surface of the fossil diesel-immersed sample (Cursaru et al. 2014)(Fazal et al. 2011b). The obtained results from FT-IR spectra matched the outcomes by XRD.

The corrosion morphologies on the surface metal could be also detected by using SEM/EDS analysis, which helped advance the understanding of the influences of biodiesel on the metal corrosiveness behaviours. The results of SEM/EDS analysis also showed the corrosion level of four types of used samples in biodiesel and fossil diesel fuel (Hu et al. 2012). The achieved results also demonstrated the corrosiveness level as follows: copper> aluminium>mild carbon steel>stainless steel. The analysis based on SEM/EDS can be found in references (Fazal et al. 2011c;Cursaru et al. 2014;Jin et al. 2015).

Corrosion may be defined as an electrochemical process brought about by a small water content contained in biodiesel. The electrochemical method is thought to be a good option since it could give results in a short time. Polarisation and electrochemical impedance spectroscopy (EIS) are usually used to determine the corrosion resistance of metals to biodiesel (Kamiński et al. 2008). In a studyby Dáaz-Ballot et al.(Díaz-Ballote et al. 2009), the electrochemical techniques, including open circuit potential (E_{ocp}) measurement, EIS, and anodic polarisation measurement were used. A piece of aluminiumwas immersed in biodiesel, and the electrochemical parameters were measured after several washing cycles. The results showed that a decrease of aluminium surface activity was due to a layer of corrosion products that was formed, resulting in a negative value, below -600 mV, of (E_{ocp}). This (E_{ocp}) value became more positive in further washing cycles (Fig.7). The above results proved that electrochemical techniques might be used to determine the corrosiveness quantitative indicator in biodiesel (Zuleta et al. 2012). This method could also be found in other studies (Santana et al. 2015;Wang et al. 2012).

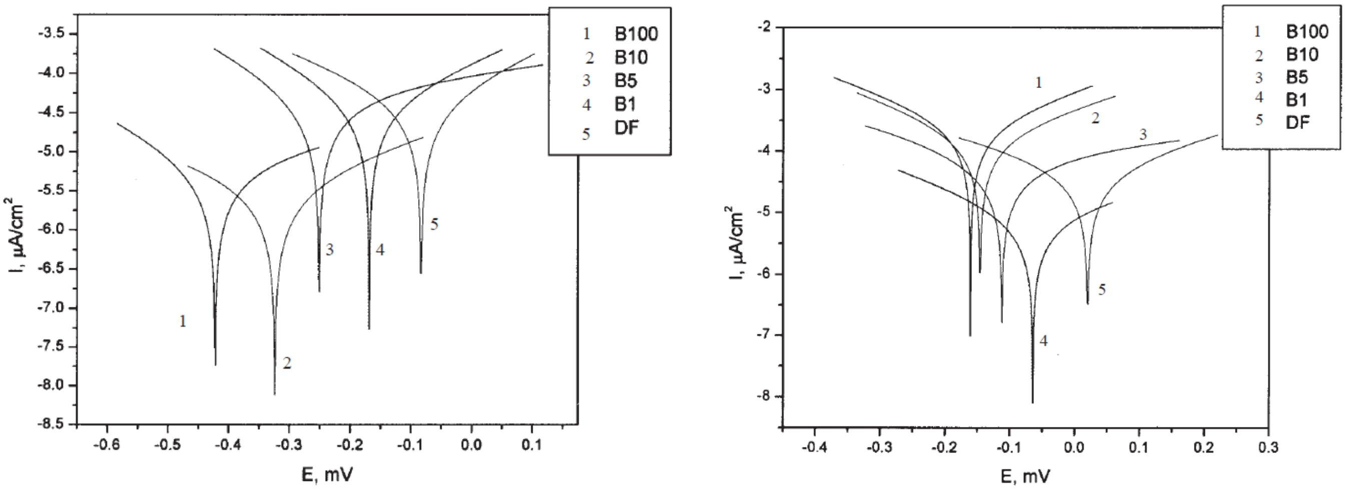


Fig. 7. The polarization curves of mild carbon steel (a), stainless steel (b) exposed to biodiesel, diesel, and blends (Diana et al. 2012)

Biodiesel was believed to be more hygroscopic in nature, possess higher conductive electrics, higher polarity and solvency than diesel fuel (Deyab 2016b). These characteristics help explain why the former may cause a higher level of corrosion than the latter. Besides, free water and oxygen content appearing in biodiesel were regarded as necessary and key materials for life, thus, and as a result a perfect environment for microbial growth (Ashraful et al. 2014). In the above process, along with the increased polarity and solvency property of biodiesel, the paints/coatings for protecting the metal could be defaced and removed. Unprotected metal could come into direct contact with biodiesel, which might lead to further chemical reactions. As a result, the complex reaction chains perpetuated the chemical or electrochemical corrosion process (Chew et al. 2013). Furthermore, due to the auto-oxidation property of biodiesel, the chemical reaction of transesterification between fatty acid and ankanol could be oxidized and reconverted into monocarboxylic acids, namely, formic acid ($HCOOH$), acetic acid (CH_3COOH), propionic acid (C_2H_5COOH), caproic acid ($C_5H_{11}COOH$) etc..., which electrolyzed H* ion, ultimately causing and enhancing the corrosion and fuel degradation (Ching et al. 2016). Moreover, Domingos et al.(Domingos et al. 2007)haveemphasised that the presence of unsaturated FAME (fatty acid methyl esters) caused thenatural oxidation in which FAME released a radical next to the double bond (C=O, or C=C), which quickly bonded with the in-air oxygen (Ragauskas 2014) afterwards. This oxidation process could have a negative impact on biodiesel properties with the increases in TAN and peroxide value being the examples, resulting in the corrosion of the components and parts, the hardening of the rubber, even the fusion of dynamical components (Sundus et al. 2017).

An experimental study was conducted by Boonyongmaneerat et al. (2011) to test the corrosiveness process in the fuel container. In the biodiesel oxidation process, due to the exposure to the air, moisture absorption and the presence of oxygen, sub-products, for example, aldehydes, alcohols, acid, and polymers, were formed along with the main products such as hydroperoxides. The appearance of shortened chain fatty acids is thought to cause an increase in the TAN values in biodiesel. From these substances, a vicious cycle starts. High TAN corrodes steel-based the container, resulting in the formation of the sediments and deposits on the engine parts, namely, injectors and pumps, or even causing the pressure to drop through the filter. A suggestion of Boonyongmaneerat et al. (2011) related to using an electrodepositingmetal aiming to protect the metal surface exposed biodiesel was presented. As a result, nickel tungsten (NiW)-based alloys were used in this study with used tungsten mass of 44% and 39% for two samples. However, only after around 2 months of testing, corrosion occurred, acid value increased by 0.3 mg KOH/g, the water content also increased by 1100ppm in comparison with the initial biodiesel components. After a long period of immersion, nickel content was found 3% in the sample with 44% tungsten, and 4% in the sample with 39% tungsten, whereas 3% tungsten content was almost constant. Factors that strongly affect the corrosive properties of biodiesel were listed in Fig.8.

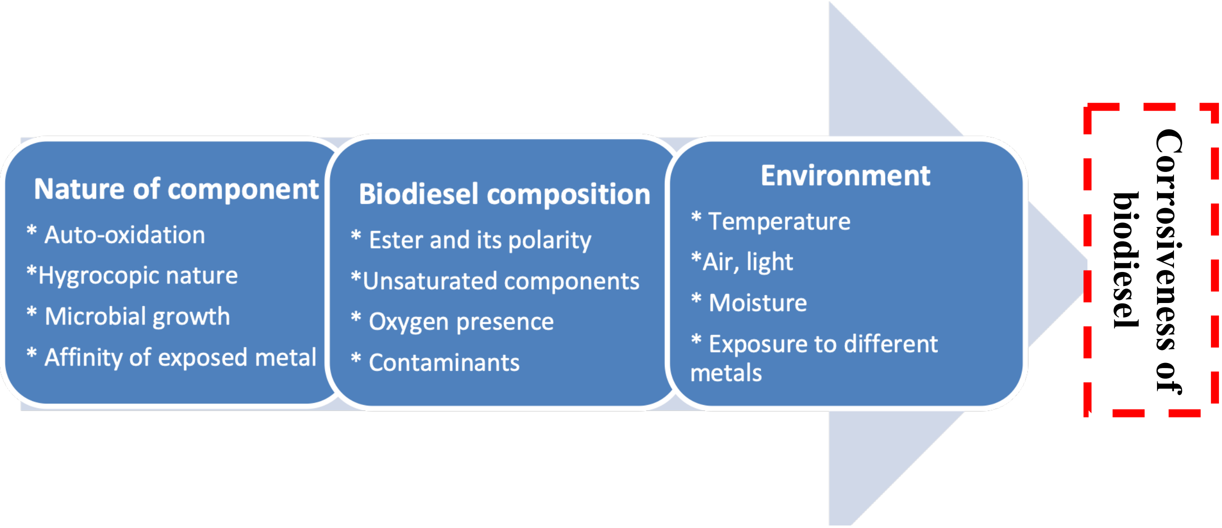


Fig. 8. Dependence of biodiesel corrosiveness on factors (Fazal, Haseeb, and Masjuki 2014)

5. Corrosion inhibitors for metals

The injurious and severe consequences of corrosion on metallic mechanical parts in the engines warrant a search for corrosion inhibitors. Some popular inhibitors include amino-amines, oxyalkylated amines, diamines, primary amines, dodecyl benzene sulfonic acids, imidazolines, naphthenic acid, phosphate esters(Jakeria et al. 2015). Corrosion inhibitors for metals in biodiesel may be different from those in fossil diesel fuel due to a gap in physicochemical properties of biodiesel (mainly mono-alkyl esters of fatty acids) compared to fossil diesel fuel (mainly hydrocarbons) (Deyab 2016). However, the inhibitors can only prolong the protection without preventing the onset of corrosion (Ashraful et al. 2014). In general, the working mechanism of the inhibitors is to form a stable metallic oxide layer that is difficult to dissolve on the metal surface being exposed to biodiesel.

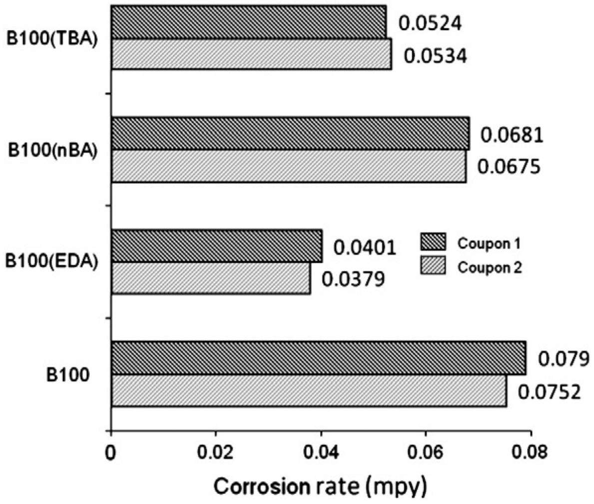


Fig. 9. The effects of inhibitors on the corrosion rates (Fazal et al. 2011b)

In a study conducted earlier, the effects of using corrosion inhibitions including dodecyl carboxylic acid and amine carboxylic acid for API 5LX steel were examined and measured by Rajasekar et al.(Rajasekar et al. 2007). The efficiency of corrosion inhibition using dodecyl carboxylic acid was 90–93%, whereas the value for amine carboxylic acid was 56–88%. In another study, isobutyl methyl-tetrahydro-azathione (IBMTAT), cyclohexyl-tetrahydro-azathione (CHTAT), cyclopentyl-tetrahydro-azathione (CPTAT) were used as corrosion inhibitors. A carbon steel sample was immersed in a solution of 20% formic acid and 20% acetic acid. The corrosion inhibition efficiencies were as follows: IBMTAT>CHTAT>CPTAT. Also, some corrosion inhibitions were found to be efficient for iron and steel in acidic media, such as isoxazolidine-based derivatives, pyridoxal hydrochloric and pyridoxol hydrochloride (Haseeb et al. 2010). Benzimidazole-2-tione, benzoxazole-2-tione were also found to be suitable for aluminium in acidic media (Bereket et al. 2004). Other corrosion inhibitors such as polyisobutylene-based succinimide derivative (SID), irganor NPA worked well to slow down corrosion in rapeseed-, and palm-based biodiesel (Hancsók et al. 2008). An antioxidant, tertbutyl hydroquinone (TBHQ), was also used in the oxidation stability correlation and corrosion on copper under SIT in the study of Almeida et al.(Almeida et al. 2011). The efficiency of oxidised reaction in converting TBHQ into tert-butylquinone (TBQ) increased the adsorption ability on the copper surface, bringing about certain protection against corrosion. The presence of TBHQ resulted in a decrease in copper concentration in biodiesel compared when there was no TBHQ. Copper concentration in biodiesel with TBHQ was only found to be 1.16μg/g after 50 hours, whereas this concentration in biodiesel without TBHQ was 3.62μg/g. A protective film layer formed on the copper surface was believed to be the working mechanism in functioning as a corrosion inhibitor. Fazal et al. carried out a study to assess the efficiency of amine-based corrosion inhibitors (Fazal et al. 2011b). Three types of theaforementioned corrosion inhibitors were chosen, namely, ethylenediamine (EDA), n- butylamine (nBA), and tert-butylamine (TBA) to evaluate and measure the corrosion resistance for the cast iron-based parts in a diesel engine. After 50 days in SIT, the corrosion rate of grey cast iron (with mass percentage of composition as follows: 3% of carbon; 1.84% of silica; 0.098% of phosphorus; 0.089% of sulfur, and iron) was 0.0771mpy, the inhibition efficiency against corrosion was found to be as follows: EDA > TBA >nBA (Fig.9). Also, the formation of oxides including FeO and $Fe_2O_3$was detected by analysing the XRD peaks. A new phase, namely, $Fe(NO_3)_3.9H_2O$, after adding TBA in biodiesel, was formed. To recap, the amine-based inhibitors created a barrier on the metal surface to prevent biodiesel or oxidation components from exposing the metal, resulting in increasing the corrosion resistance.

6. Conclusions

At present, pure biodiesel fuel is not used for commercial purposes. This may be due to some concerns regarding the failures of diesel engine components resulting from unforeseeable causes. Biodiesel can enhance the corrosion level of various parts in diesel engines, especially metal parts, as they are exposed to the fuel. Understanding the mechanism and preventing the negative effects of corrosion on mechanical parts in diesel engines in biodiesel is a big challenge for scientists, engineers and manufacturers. In this review, the higher corrosion rates of metals in biodiesel in comparison with fossil diesel fuel were demonstrated. The factors affecting corrosion level and modes such as component nature, biodiesel composition and environment were also reported. In diesel engines, commonly-used metals and alloys were copper, aluminium, and steel. They were believed to be easily prone to corrosion in biodiesel. In the static immersion test, pitting corrosion was considered to be common with nonferrous metals and alloys, and carbon steel.

Nevertheless, stainless steel seemed to be immune to pitting corrosion. Based on the published results, the corrosion property of copper was the highest, followed by that of aluminium. As for the level of corrosion resistance, carbon steel was found to be lower than stainless steel and higher than aluminium. Finally, corrosion inhibitors added to biodiesel aiming at delaying the onset of corrosion were also introduced.

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