Full Paper

CORROSION BEHAVIOUR OF BRASS, GALVANIZED STEEL AND STAINLESS STEEL IN BLENDS OF JATROPHA BIODIESEL AND DIESEL

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ABSTRACT

The corrosion behaviour of brass (Br), galvanized steel (GS) and stainless steel (SS) on exposure to Jatropha biodiesel and its blend with petroleum–based diesel was investigated in this study. Jatropha biodiesel was produced via transesterification reaction with methanol using sodium hydroxide as catalyst. Corrosion rates of these metals were evaluated using weight loss method after the metals were immersed in the fuels for 8 weeks. The acid value (AV), peroxide value (PV) and iodine value (IV) of the fuels during the immersion test were analysed. The degradation of the metal surfaces and the colour of the biodiesel fuel at the end of the immersion test were also characterized by visual inspection. The corrosion rate of Br, GS and SS in Jatropha biodiesel after 8 weeks (56 days) of exposure was 0.6091, 0.3435 and 0.053 mpy respectively. As the volume of diesel was increased in the blend, the corrosion rates of the alloys decreased. There was increase in both the AV and PV during the immersion test with the fuel containing Br having the highest value. A decrease was observed in the IV of the Jatropha biodiesel during the immersion period. Changes in colour were also observed for the biodiesel containing the metals.

Keywords: *Jatropha biodiesel, Corrosion, Acid Value, Stainless Steel, Galvanized Steel, Brass, Fuel*

1. INTRODUCTION

Petroleum based fuel became a primary source of energy for transportation from the 20th century. This has continued till the beginning of the 21st century with all vehicles running on gasoline, diesel or natural gas. Currently, due to gradual depletion of world petroleum reserves and its impact on the environment as a result of increased exhaust emission, there is an urgent need to develop an alternative source of energy for transportation. Biodiesel which is a fitting alternative to petroleum based diesel fuel is one of such fuel that can be used as an alternative to diesel fuel (Knothe, 2005). Biodiesel is a domestically renewable fuel that is used in compression ignition engine. It is produced from the reaction of either vegetable oils or animal fats with an alcohol in the presence of a catalyst. Biodiesel has several advantages over regular petroleum diesel in that biodiesel produces significantly less harmful emissions than petroleum diesel when burned in a compression ignition engine. It is also non-flammable (high flash point), and biodegradable. However, one of the drawbacks of biodiesel usage is its oxidation which is due to its chemical composition (Knothe *et al.*, 1997). Biodiesel comprised of unsaturated fatty acid methyl ester molecules which easily undergo oxidation during storage. Also, biodiesel tends to have a more corrosive effect than regular diesel on metals (Fazal *et al.*, 2010). Corrosion is the disintegration of a material as it interacts with the environments where it finds itself. According to Nernst's theory, all metals have a tendency to pass into solution (Fontana, 1987). However, the extent of corrosion varies for metal ions depending on its oxidation potential and various prevailing conditions such as temperature, water content, etc. in the fuel. The level of corrosion also depends on the type of metal in contact with biodiesel fuel. (Agarwal, 2007). Compared with diesel, biodiesel is more prone to absorb water; this is because the esters in biodiesel exhibit some hygroscopic properties thereby increasing the water content. The water tends to condense on the metal surface causing corrosion and deterioration of the material. Furthermore, the corrosion of metallic parts in contact with biodiesel fuel over a long period of time can cause fuel degradation, thereby lowering the quality of the fuel. Properties like the acid value, peroxide value and viscosity increases, while the iodine value decreases. All of these changes affect the quality of biodiesel (Haseeb *et al.*, 2010; Wang *et al.*, 2011). Several researchers have studied the corrosive effect of various biodiesel on the corrosion rates of metals. Hu *et al.* (2012) investigated the corrosion behaviours of metals in biodiesel from rapeseed oil and methanol. Their results showed that the corrosion rates of copper, mild carbon steel, aluminium and stainless steel in the biodiesel were 0.02334, 0.01819, 0.00234 and 0.00087 mpy respectively. In an earlier study conducted by Diaz – Ballote *et al.* (2009), the corrosion behaviour of aluminium exposed to canola oil biodiesel was investigated. They found that the corrosion behaviour of aluminium in biodiesel contaminated with alkalis is similar to the corrosion behaviour of aluminium in aqueous solution. The presence of metals has also been found to have effect on the properties of biodiesel. The results obtained by Fazal *et al.* (2010) revealed that palm biodiesel exposed to metals shows significant degradation in fuel properties as evidence by increase in acid number, viscosity and density at the end of the immersion test. In another study carried out by Fazal *et al.* (2011), they found out that there were increases in acid number and density of palm biodiesel exposed to mild steel at the end of the immersion test. Chew *et al.* (2013) also observed increase in the total acid number of palm biodiesel exposed to aluminium and magnesium at the end of the immersion test. However, the changes in these physicochemical properties of biodiesel exposed to metals over time have not been fully reported. It is therefore imperative to study the changes in properties of Jatropha biodiesel exposed to metals over time.

The aim of this present study therefore was to investigate the corrosion behaviour of brass, stainless steel and galvanized steel in Jatropha biodiesel and its blends with petroleum diesel. Cast iron, galvanized steel and brass are common metals that can be used as storage systems for biodiesel, while copper, low mild carbon steel, aluminium and stainless steel are mostly used in the manufacture of automobile transmission component parts that used diesel as fuel. The presence of metals on some physico-chemical properties in addition to the colour change of Jatropha biodiesel was also investigated in this study.

2. MATERIALS AND METHODS

2.1. Materials

Jatropha seed oil was obtained from a local market in Auchi, Edo State, Nigeria. Petroleum diesel was purchased from a local petrol filling station in Benin City, Edo State, Nigeria. Galvanized steel (81.75% Fe, 18% Zn and 0.25% C), 18/8 stainless steel (73.85 % Fe, 18% Cr, 8% Ni and 0.15% C) and brass (63.7% Cu and 36.3% Zn) was obtained from the Faculty of Engineering Workshop of the University of Benin, Nigeria. All chemicals (anhydrous Methanol of 99.95% purity, sodium hydroxide pellet, ethanol, benzene, chloroform, glacial acetic acid, potassium hydroxide, potassium iodide, sodium thiosulphate, Wij's reagent and acetone) used were of analytical grade and were obtained from Stanvac Laboratory in Benin City, Nigeria.

2.2. Jatropha Biodiesel Synthesis and Physico-chemical Analysis

The initial free fatty acid content of the Jatropha seed oil was 8%, thus a two–step transesterification process as described by Sarin *et al.* (2010) was used for the production of the biodiesel. The physico-chemical properties of the synthetized Jatropha biodiesel and the diesel were determined according to ASTM and AOCS methods.

2.3. Corrosion Experiment

The corrosion of SS, GS and Br in blends of Jatropha biodiesel and petroleum based diesel were investigated by static immersion test. The tests were carried out at room temperature for eight weeks. The metal coupons used were machined and polished into various strips of SS (21 x 16 x 0.9 mm), GS (21 x 16 x 1.1) and Br (21 x 16 x 1.55 mm). A hole of 2 mm was drilled at one end of each metal coupon for hanging the test coupons in the fuel. Before the metals were immersed, they were polished with silicon abrasive paper (320 grit) after which they were cleaned with distilled water and detergent. Acetone was used to swab the metal coupons and a hand dryer was then used to dry them. A total of twelve metal coupons (comprise of four each for SS, GS and Br) were used for this study. One metal coupon was immersed in a glass beaker containing 50 cm³ of B100 (Jatropha biodiesel), B20 (diesel containing 20 vol. % biodiesel), B5 (diesel containing 5 vol. % biodiesel) or B0 (petroleum diesel) fuel. The beaker was covered and placed on a table. At the end of the immersion test, the coupons were removed from the beaker and each coupon was degreased by placing it in acetone. The coupons were then scrubbed with soft silicon abrasive paper and a polymer brush was used in a stream of water to remove the corrosion products. The coupons were further placed in acetone and a hand dryer was used to dry the coupon. The weight of the test coupons were measured by an electronic weighing balance (Pioneer, Model PA Z13, OHAUS Corp.) with $\pm$ 0.001g accuracy before and after immersion. The weight loss after immersion was converted into corrosion rate in mils per year (mpy) using Eq. 1. (Fontana, 1987).

$$\text{Corrosion rate (mpy)} = \frac{534 \times w}{D \times A \times t} \quad (1)$$

where corrosion rate, mpy stands for mils (0.001 inch) per year, w is the difference in weight of the test coupons before and after immersion in mg, D is the density of the metal in g/cm³, A is the exposed surface area of the coupon in square inch and t is the exposure time in hours. In this study, the densities of the metals used were 7.9, 7.85 and 8.4 g/cm³ for SS, GS and Br respectively.

Changes in surface appearance of the metal coupons and colour of the fuel were carried out by taking photographs of the samples before and after the immersion test using a SONY Digital Still Camera (DSC – W310). Physico-chemical analyses of fuels were done at intervals of two weeks during the immersion. The measurement of the density of the fuel was done according to the ASTM D1298 standard, while the total acid number (TAN) and peroxide number (PV) were analysed according to ASTM standard D664 and AOAC standards (Firestone, 1998) respectively. The iodine value (IV) was measured using the American Oil Chemist's Society method (AOCS, 1997).

3. RESULTS AND DISCUSSION

3.1. Physico-chemical Properties of Jatropha Biodiesel and Diesel

The physico-chemical properties of the Jatropha biodiesel and petroleum based diesel in addition to the ASTM D6751 standard for biodiesel are shown in Table 1. The density of the freshly produced Jatropha biodiesel was 0.8760 g/cm³. The value obtained for the density falls within the ASTM D6751 range from 0.860 to 0.90 g/cm³.The acid value obtained for the produced biodiesel was 0.4165 mg KOH/g which was within the range of ASTM value of less than 0.8 mg KOH/g.

Table 1. Physico-chemical properties of Jatropha biodiesel and diesel

Property D6751	Unit	Jatropha Biodiesel	Petroleum Diesel	Test method ASTM	ASTM Limit
Density (20°C)	g/cm³	0.8740	0.840	D1298	0.86 – 0.90
Acid value	mg KOH/g	0.4165	0.140	D664	0.8 max
Viscosity (40°C)	mm²/s	1.904	N/d	D445	1.9 - 6.0
Flash point	°C	136	74	D93	130 min
Cetane number	-	51	52.1	D613	47 min
Peroxide value	meq.O_2/kg	0.50	N/d	AOCS	-
Iodine value	mg I_2/100g	98.525	N/d	AOCS	120 min

N/d: Not determined

The viscosity of the biodiesel was calculated to be 1.904 mm²/s which was also within the ASTM D6751 standard of between 1.9 and 6.0 mm²/s. The flash point of the produced biodiesel was 136 °C which was higher than the minimum value of 130 for the ASTM standard. The iodine value (IV) for the freshly produced biodiesel was 98.525 g I_2/ 100g. The cetane number obtained was 51 which was also higher than the minimum requirement for biodiesel. Although the peroxide value (PV) is not specified in the biodiesel standard, it has an influence on the cetane number. The PV obtained for the biodiesel was 0.50 meq.O_2/kg.

3.2. Peroxide Value of Jatropha Biodiesel during the Immersion Test

The peroxide value (PV) gives a measure of the primary oxidation products that are formed during the oxidation of biodiesel (Pullen and Saeed, 2012). These products are mainly peroxides and hydroperoxides. The changes in the PV of Jatropha biodiesel containing the different metal coupons are shown in Fig. 1. Before the immersion test, the PV of the biodiesel was 0.5 meq.O_2/kg. However, there was increase in PV for all biodiesel samples as the immersion time increases.

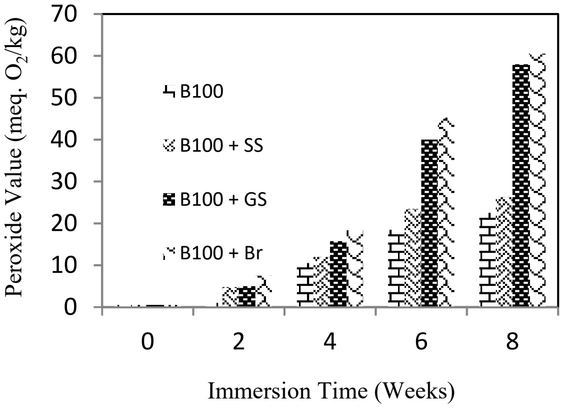

Fig. 1. Changes in the Peroxide Value of Jatropha Biodiesel (B100) during the Immersion Test at room temperature

The biodiesel containing Br had the highest PV of 60.53 meq. O_2/kg at the end of the immersion period. This was followed by the biodiesel containing GS and SS with a value of 58 and 26.28 meq O_2/kg respectively. The biodiesel sample without metal had the least PV of 22.5 meq. O_2/kg at the end of the immersion period of 8 weeks. Increase in PV of biodiesel containing metals has also been reported by several other researchers. Fernandes *et al.* (2013) observed increased in PV of soybean biodiesel over time in contact with carbon steel. Akhabue *et al.* (2014) also observed increase in PV of castor oil biodiesel containing iron and aluminium contaminants. The presence of metals has been found to increase the oxidation of biodiesel. The primary oxidation products that are formed during the oxidation of biodiesels are mainly peroxides and hydroperoxides. Copper and brass (a copper alloy) is known to be essentially the worst offender. So the increase in the PV of Jatropha biodiesel in which the metals were immersed was not unusual.

3.3. Iodine Value of Jatropha Biodiesel during the Immersion Test

The iodine value (IV) for biodiesel gives a measure of the degree of unsaturation of the biodiesel fuel. The higher the IV, the more the degree of unsaturation of the biodiesel fuel. The changes in the IV of the biodiesel during the immersion period are shown in Fig. 2. The figure shows that the IV for the biodiesel decreased with increase in immersion time. This is probably as a result of the fact that, as the immersion time increases, the degree of unsaturation of the biodiesel fuel decreases due to oxidation.

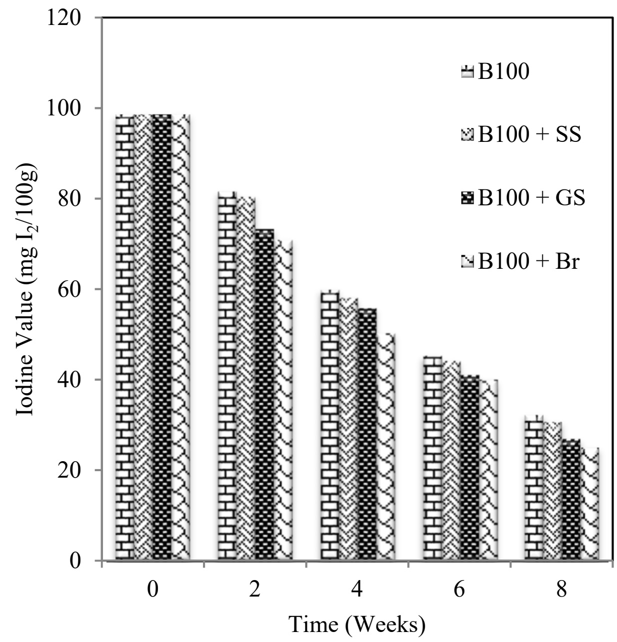

Fig. 2. Changes in the Iodine Value of Jatropha Biodiesel during the Immersion Test at room temperature

The oxidation of the biodiesel over time caused the breakdown of the carbon double bonds in the fuel. This causes the degradation of the fuel (Pullen and Saeed, 2012). Since the biodiesel containing SS were the least oxidized, they had higher degree of unsaturation and thus higher IV, while the biodiesel containing Br had the least IV and biodiesel containing GS had IV greater than those containing Br but lower than those of containing SS.

3.4. Acid Value of the Jatropha Biodiesel and its Blends during the Immersion Test

The changes in the acid value (AV) for the fuels containing the different metals are shown in Fig. 3. The AV of biodiesel gives an indication of the extent of oxidation the fuel has undergone. Acids are secondary oxidation products formed during the oxidation of biodiesels (Pullen and Saeed, 2012). There were significant increases in the AV for all the fuels during the 8 weeks period of investigation. As seen in Fig. 3a, the AV of the Jatropha biodiesel (B100) containing Br increased from an initial value of 0.4165 mg KOH/g to 2.5245 mg KOH/g during the 8 week of immersion. The AV of the fuel containing SS was 1.503 mg KOH/g after the same period. The changes in the AV for the B20 fuel containing the three different metals are shown in Fig. 3b. It was also observed that the fuel containing Br had the highest AV at the end of the immersion test. This was followed by the fuel containing GS and SS in that order. Similar trend was observed for the Jatropha biodiesel blend (B5) and diesel (B0) as shown in Fig. 3c and 3d respectively. It was also observed that the fuel samples that had no metal immersed in them had the least AV after the period of investigation. This is an indication that oxidation of biodiesel is increased in the presence of metal. In all, it was observed that the AV of the fuels for any of the fuel containing a particular metal type decreased in the order: B100 › B20 › B5 › B0. This is as a result of the fact that the secondary oxidation products of biodiesel are mainly acids. It therefore implies that as the as the ratio of Jatropha biodiesel to petroleum based diesel increases, so does the acid value. It can be seen that the acid values for all the

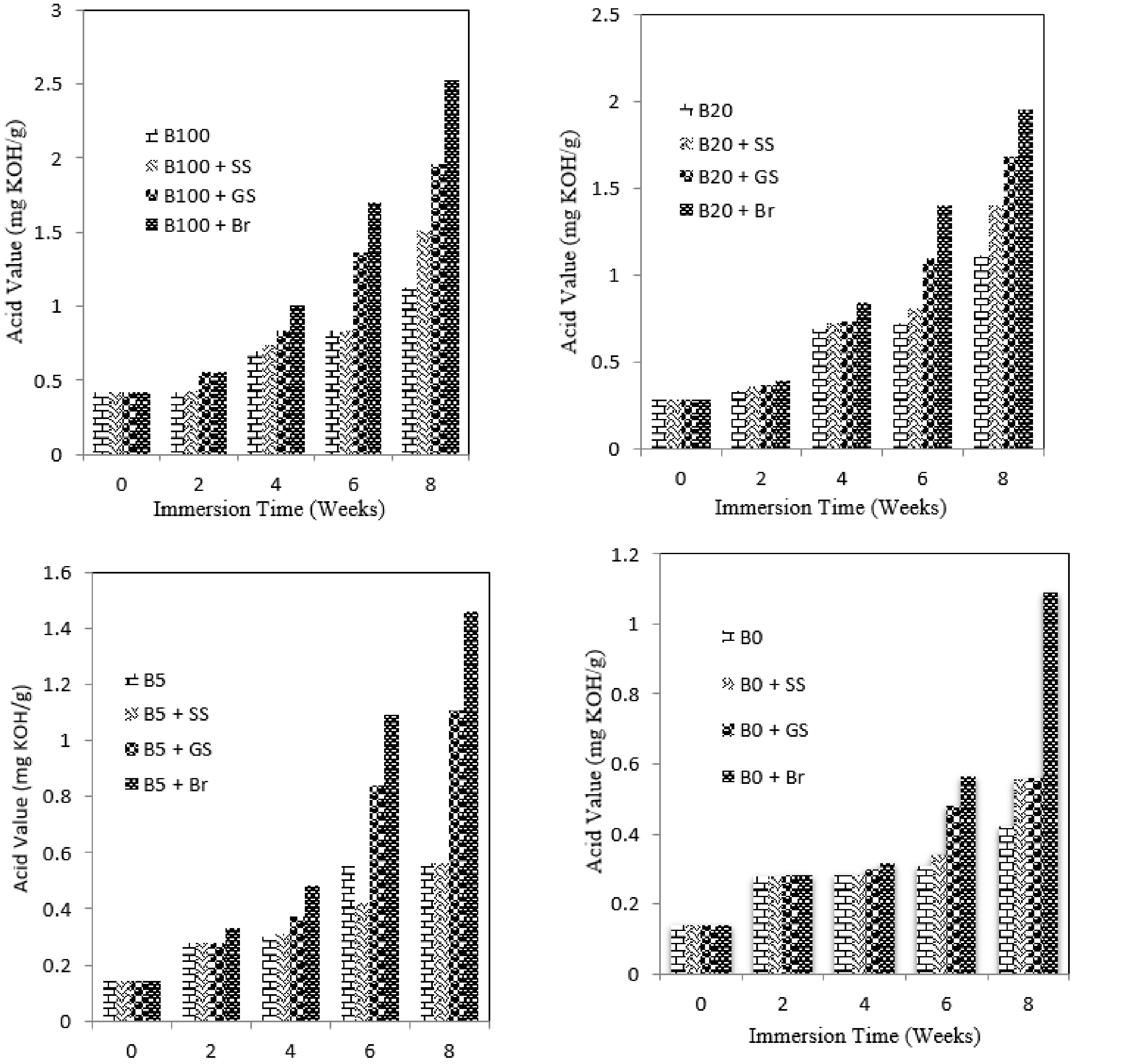

Fig. 3. Changes in Acid Value of the different fuels during the immersion test at room temperature (a) B100 (b) B20 (c) B5 (d) B0

fuels for any metal type increases with increase in immersion time of metal in the fuel. After production of fuels, the acid value for both petroleum based diesel (0.1375 mg KOH/g) and biodiesel (0.4165 mg KOH/g) were within acceptable ASTM standards of 0.5mg KOH/ g of fuel, but after 2 weeks, only values for stainless steel exposed biodiesel were within range, those of brass and galvanized steel were above the standard limit. The diesel samples were within accepted limits until after 8 weeks for stainless steel and galvanized steel and 6 weeks for brass.

From the results, it was noticed that fuels exposed to Br had the highest acid value in (B100, B20, B5 and B0) than fuels exposed to the other two metals. The AV of the biodiesel exposed to SS was lower than that exposed to GS. This should be expected since metals with higher corrosion rates deposits ions in the fuels. These ions enhance the corrosion process which leads to an increase in the acid content of the biodiesel fuel. Brass has been found to be more corrosive in biodiesel compared to other metals (Fazal *et al*., 2012). It has been reported that Br containing 15% Zn is susceptible to dezincification. Zinc has a high tendency to dissolve into solution, whereas Cu has a high tendency to plate. This is as a result of the fact that the electrochemical potential for Zn is -0.763 V, whereas that for Cu is 0.337 V. The presence of zinc ions (metallic ions) in the biodiesel will catalysed the oxidation of the biodiesel (Pullen and Saeed, 2012) The implication is that the biodiesel fuel in which Br was immersed will have a higher AV, and the biodiesel in which stainless steel was immersed will have the least acid values because stainless steel is the least corrosive of the three meta

3.5. Corrosion Rate

The corrosion rates of the metals in Jatropha biodiesel and its blend with diesel after immersion at room temperature (28 ± 2 °C) for 8 weeks (1334 h) are shown in Fig. 4. From the figure, it is seen that for each of the metal investigated, the corrosion rate was greater in the Jatropha biodiesel fuel (B100) than the Jatropha biodiesel blend (B20 and B5) and diesel (B0). The corrosion rate was found to decrease in the order: B100 › B20 › B5 › B0. The increase in corrosion rate of the different metals in Jatropha biodiesel as compared to that in diesel (B0), B20 and B5 could be attributed to the presence of oxygen and moisture absorption. Compositionally, biodiesel contains 10 - 12 wt. % oxygen, while diesel contains no oxygen. In the presence of moisture and oxygen, metals could easily have oxidized to different metal oxides and later it forms different metal compounds by further oxidation (Fazal *et al.* 2012). This is why in most cases, biodiesel exposed metal surface shows higher oxygenated species (Fazal *et al.*, 2011; Chew *et al.*, 2013; Fazal *et al.*, 2013; Cursaru *et al.* 2014). In addition, the ester molecules of biodiesel are more hygroscopic and polar in nature as compared to diesel. As the percentage of biodiesel increases in the blend, so also does its tendency to absorb more water and be oxidized. It was also observed that, of the three metals investigated; Br is more corrosive followed by GS while SS exhibited the least corrosivity. The corrosion rate of the metals in biodiesel (B100) was 0.6091, 0.3435 and 0.053 mpy for Br, GS and SS respectively.

The high corrosion rate of Br in biodiesel has been reported by several other researchers. The corrosion of Br occurs by dezincification. Dezincification is a form of de-alloying in which Zn separates by dissolution from Br as shown in Eq. (2) (Ahmad, 2006).

$$Zn \rightarrow Zn^{2+} + 2e \quad (2)$$

The corrosion rate of the metals in the other fuels follows the same order as the corrosion rate of Br was greater than GS and SS.

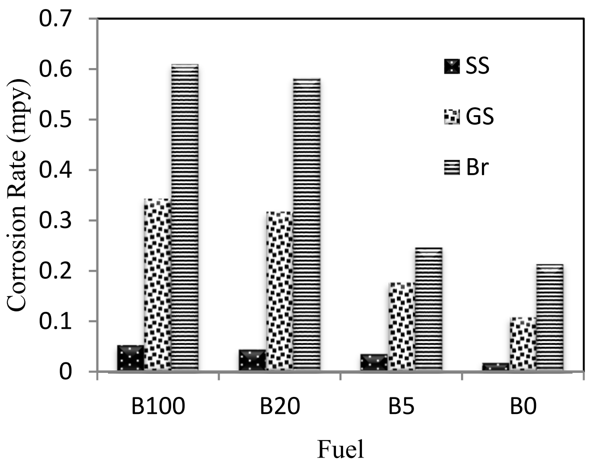

Fig. 4. Corrosion Rates of Stainless Steel, Galvanized Steel and Brass in the Blends of Jatropha Biodiesel and Diesel.

3.6. Surface Appearance of the Metal

The surface appearance of the different metal coupons before and after exposure to Jatropha biodiesel and diesel are shown in Fig 5. It was observed that as-received, Br is brown, GS and SS are ash in colour as seen in Fig. 5a. After exposure to biodiesel for 8 weeks, patches of green layer were formed on Br as shown in Fig. 5b. The green layer of corrosion products are most likely be copper carbonate ($CuCO_3$). The reactions which lead to the formation of $CuCO_3$ are shown in Eqs. (3 - 6). Fazal *et* al. (2012) reported the presence of $CuCO_3$ in copper based alloy immersed in palm biodiesel. It was also observed that reddish brown layers were also formed on the Br exposed to diesel as shown in Fig. 5c. The reddish brown layer may probably be cuprite oxide

$$2Cu + O_2 \rightarrow 2CuO \quad (3)$$
$$4Cu + O_2 \rightarrow Cu_2O \quad (4)$$
$$2Cu_2O + 4CO_2 + O_2 \rightarrow 4CuCO_3 \quad (5)$$
$$CuO + CO_2 \rightarrow CuCO_3 \quad (6)$$

For GS, a reddish brown layer which may be Fe_2O_3 was also formed after exposure in Jatropha biodiesel and diesel as shown in Fig. 5b and 5c. The reaction leading to the formation of Fe_2O_3 is shown in Eq. (7). Cursaru *et* al. (2014) reported the presence of Fe_2O_3 on cast iron exposed to sunflower biodiesel and diesel. There was no noticeable change in the surface appearance of SS immersed in biodiesel and diesel after the 8 weeks period of immersion.

$$4Fe + 3O_2 \rightarrow 2Fe_2O_3 \quad (7)$$

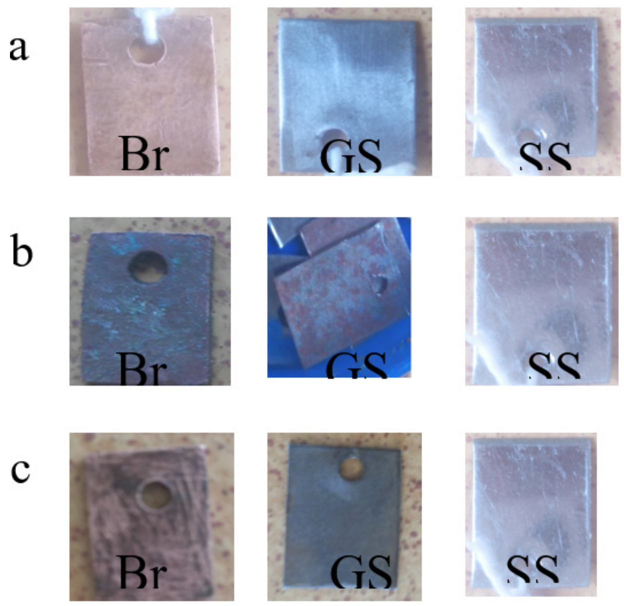

Fig. 5. Photographs of the metals (a) before immersion and after immersion in (b) Jatropha biodiesel (B100) (c) diesel (B0)

3.7. Colour of Biodiesel Fuel

The colour of the biodiesel before and after exposure to the different metals is shown in Fig. 6. It is seen from Fig. 6a, that the freshly produced biodiesel has a cloudy milk colour before the immersion test. After exposure of the biodiesel to stainless steel, galvanized steel and brass, the colour of brass-exposed biodiesel was changed from a cloudy milk colour to a yellowish green clear colour as seen in Fig. 6b. The yellowish green colour is probably due to the presence of ZnO as seen in Eq. (8). Zinc oxide is formed as a result of the corrosion of Br immersed in biodiesel (Fazal *et al.*, 2012). Fazal *et al.* (2012) has also reported the presence of Cu_2O, CuO and $Cu(OH)_2$ in biodiesel exposed to brass (Eqs. (9 and 10)). The biodiesel exposed to GS changed from the cloudy milk colour to clear reddish brown colour as shown in Fig. 6c. The colour change may be attributed to the presence of $Fe(OH)_2$ as shown in Eq. (11) (Cursaru *et al.*, 2014). The biodiesel exposed to stainless steel became clear and transparent after 8 weeks (Fig, 6d). Fig. 6e **shows the colour of the biodiesel in which no metal has been added.** The colour of the biodiesel became transparent from the initial cloudy milky colour that was observed before the immersion test.

Fig. 6. Colour of Jatropha biodiesel (B100) (a) after production and after exposure to (b) brass (Br) (c) galvanized steel (GS) (d) stainless steel (SS), (e) without metal after eight weeks

$$2Zn + O_2 \rightarrow 2ZnO \quad (8)$$
$$2Cu + O_2 + 2H_2O \rightarrow 2Cu(OH)_2 \quad (9)$$
$$CuO + 2H_2O \rightarrow 2Cu(OH)_2 \quad (10)$$
$$4Fe + 2O_2 + 4H_2O \rightarrow 4Fe(OH)_2 \quad (11)$$

4. CONCLUSION

Corrosion rate and degradation of brass, galvanized steel and stainless steel in Jatropha biodiesel and its blends with diesel was investigated by static immersion test. The following conclusions were drawn from the study:

i. For the three metals investigated, plain Jatropha biodiesel is more corrosive than the petroleum based diesel. The Jatropha biodiesel is also more corrosive than the blends of Jatropha biodiesel mixed with petroleum-based diesel.
ii. Upon exposure to Jatropha biodiesel, the corrosion rate for the three metals under investigation is 0.6091, 0.3435 and 0.053 mpy for Br, GS and SS respectively.
iii. Brass is less resistant to corrosion in biodiesel and their blend with petroleum diesel compared to stainless steel and galvanized steel and causes more degradation to fuel properties as measured by changes in PV, IV and AV.
iv. As time of exposure of the metals to Jatropha biodiesel increases, AV and PV properties of the biodiesel increase, while IV decreases.
v. The degradation of the surface of each metal exposed to biodiesel is higher than that exposed to petroleum based diesel.
vi. There were varying changes in the colour of the biodiesel exposed to the different metals at the end of the immersion test.

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