

Evaluation on Physical and Chemical Properties of Aviation Biofuels from Corn Straw Oil by Hydrothermal Liquefaction Based on Chemical-Biological Pretreatment

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This study extended previous work investigating aviation fuels and a 1:1 (by volume) blend of those fuel with a biofuel derived from corn straw oil prepared by the hydrothermal liquefaction of chemical-biological pretreated. The results showed that the aviation kerosene based on biomass and aviation gasoline based on biomass exhibited 3 and 0.11 MJ/kg higher heating values, 53.6- and 1.5-fold higher total acid numbers, 14.4 % and 20.7 % lower freezing points, and 62- and 123-fold higher existent gum contents, respectively. Bio-oil from hydrothermal liquefaction based on chemical-biological pretreatment could be compatible with aviation kerosene by esterification or catalytic hydrogenation.

1. Introduction

In recent years, the rapid development of the aviation industry has led to sudden increases in the consumption of aviation fuel, resulting in elevated emissions of greenhouse gases. The consumption of aviation fuel accounts for 2 % of the total consumption of fossil fuels, equivalent to 12 % of the fuel consumption in the transportation sector. However, many challenges remain before aviation biofuels can be widely adopted, particularly with regard to cost and sustainability. A biomass-based fuel has proved to have the great potential to replace petroleum-derived aviation fuels [9].

At present, biofuel blending substitute conventional aviation fuels for aviation biofuels is the most potential method [7]. Therefore, both the production of bio-oil by using stalk and the upgrading of the aviation biofuels utilization technologies have attracted much research attention in recent years [10]. The key to achieving this goal is a technology through which the stalk can be liquefied to bio-oil [1]. Sub-Supercritical Fluid Technology can enhance the mass transfer between stalk and the solvent [3]. Additionally, the reaction time is shorter than the traditional pyrolysis reaction, the residue and the gas produced are little in quantity, the efficiency is high while the cost is low. Researchers currently either use water or an organic solvent as a medium. Easy availability, low price and potential for industrial application are the characteristics which make water a suitable choice for being the reaction medium. Due to strong dissolution abilities for reactants and products, ethanol is the most favoured medium among organic compounds. However, if the bio-oil is used as an alternative fuel, ethanol, which is used as a medium, does not need to be excluded from the product. Production of bio-oil from water and alcohol by using eco-friendly technology of Sub- and Supercritical fluid has attracted much research attention in recent years. Utilizing bio-oil from the hydrothermal liquefaction of lignocellulose straw to synthesize aviation biofuels has become a focus of investigation, but the complex structures and range of constituents of lignocellulose have become obstacles to improving the yield and quality of the bio-oil.

Previous studies by our group [5] have shown that pretreating corn straw using acid-alkali solution and yeast (called chemical-biological pretreatment) could destroy the lignin structure and expose the bare cellulose and hemicelluloses to acid solution for conversion into sugars. During the fermentation process, the sugars are consumed by the yeast to produce ethanol, a potential material of hydrothermal liquefaction. Preparing bio-oil by the hydrothermal liquefaction of chemical-biological pretreated corn straw could decrease the variety of chemical constituents of the bio-oil and increase the yield of bio-oil while enhancing its content of chemical constituents that possess qualities of antidetonation and inoxidisability, such as toluene, 4-hydroxy-4-methyl-2-pentanone, and 2,6-di-tert-butyl-4-methylphenol, enabling its potential as an excellent biofuel.

Aviation biofuels were prepared by directly mixing aviation fuels with bio-oil from the hydrothermal liquefaction of chemical-biological pretreated corn straw in this paper. Over the course of this work the compatibility of bio-oil with aviation fuels was assessed by analysing the physical and chemical properties of aviation biofuels, which aims to provide the corresponding guidance for preparation and upgrading of bio-oil.

2. Materials and methods

2.1. Materials and reagents

RP-3 aviation kerosene and NO.95 aviation gasoline were provided by a petrochemical company, and their major specifications were shown in Table 1. Chemical-biological pretreated corn straw was the material for the preparation of the bio-oil.

Table 1: Major specifications of RP-3 aviation kerosene and NO.95 aviation gasoline

Items	Unit	RP-3 aviation kerosene	NO.95 aviation gasoline
Total acid number	mg KOH/g	≤ 0.015	≤ 1
Initial boiling point	°C	147	≥ 40
10 % distillate temperature		≤ 205	≤ 80
50 % distillate temperature		≤ 232	≤ 105
90 % distillate temperature		218	≤ 145
End point of distillation		≤ 300	≤ 180
Residual volume fraction	%	≤ 1.5	≤ 1.5
Loss volume fraction		≤ 1.5	≤ 1.5
Octane rating		Nr	95
Existent gum	mg/100 ml	≤ 7	≤ 3
Flash point	°C	≥ 38	Nr
Freezing point	°C	≤ -47	≤ -58
Kinematic viscosity at 20 °C	mm ² /s	≥ 1.25	Nr
Lower heating value	MJ/kg	≥ 42.8	≥ 43.5
Density at 20 °C	kg/m ³	775 ~ 830	748
Copper corrosion 2 h at 100 °C		≤ 1	≤ 1
Saturated vapor pressure	Kpa	Nr	27 ~ 48
Sulfur content	wt. %	≤ 0.2	≤ 0.05
Aromatics content	%, V/V	≤ 20	≤ 35

Nr: not required

Based on our previous research [5], the autoclave was heated to the desired temperature (370 °C) for 30 minutes by a heating jacket. After the reaction, the autoclave was cooled to 25 °C by cool water, and the gas was exhausted via an exhaust valve. The collection and separation process of the products were shown in Figure 1. Figure 2 and Table 2 showed the chemical compositions of bio-oil.

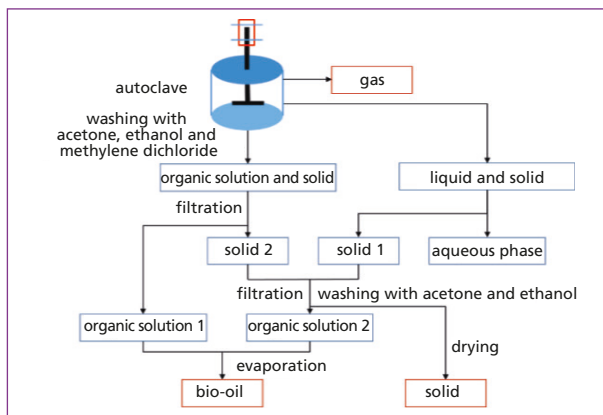


Figure 1:

Collection and separation procedure of liquefaction products

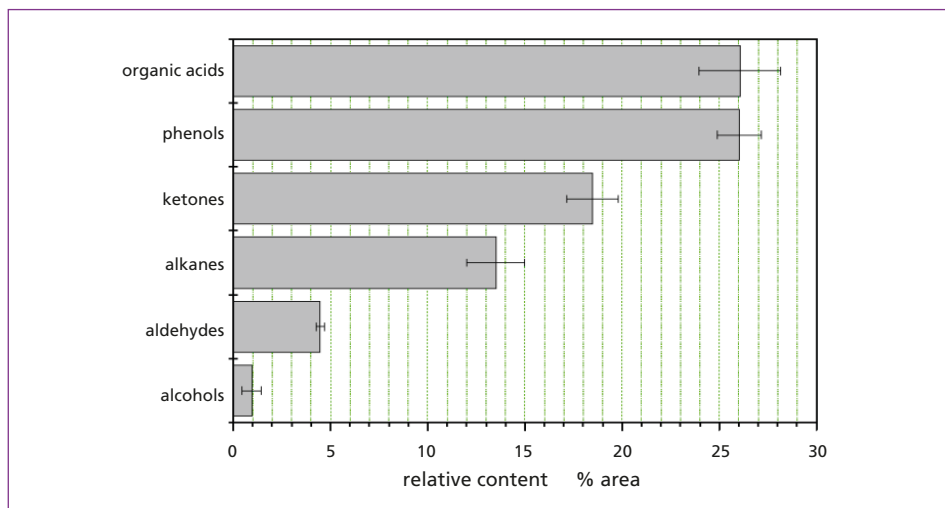


Figure 2: Relative content of several compounds in bio-oil

Table 2: Main components of bio-oil

No.	Compounds	RT min	Relative content % area
1	Ethanol	1.355	0.95
2	Toluene	2.764	0.65
3	Cyclopentanone	3.060	0.71
4	2-Methylcyclopentanone	3.815	0.79
5	Diacetone alcohol	3.858	1.67
6	3-Methylcyclopentanone	3.936	0.69
7	Methyl cyclopentenolone	5.036	2.11
8	(Z)-4-Decenal	5.586	0.55
9	3-Methyl-2-cyclopenten-1-one	6.190	1.78
10	Phenol	7.114	2.12
11	Thujone	7.199	1.77
12	2,3-Dimethyl-2-cyclopentenone	7.495	5.21
13	trans,trans-2,4-Heptadienal	8.196	1.05
14	Cinnamaldehyde	8.728	3.92
15	4-Ethylphenol	10.075	2.47
16	4-Ethyl-2-methoxyphenol	11.422	19.99
17	1-Indanone	11.513	3.12
18	3,3-Dimethyl-1-indanone	11.833	2.30
19	2,6-Di-tert-butyl-4-methylphenol	14.431	1.43
20	Palmitic acid	19.416	26.06
21	Octacosane	22.249	3.37
22	Nonadecane	23.071	1.90
23	Pentacosane	23.875	3.70
24	Tetratetracontane	24.648	3.04
25	Heptacosane	25.403	1.49
Total area	/	/	92.84

2.2. Measurement methods and instruments

The chemical compositions of the bio-oil, the aviation biofuels and the conventional aviation fuels were analysed by a gas chromatography–mass spectrometry (GC/MS, Agilent 6890N/5975) [5].

The net heating values of aviation biofuels and conventional aviation fuels were measured in accordance with ASTM D240 using an oxygen bomb calorimeter. The freezing points were measured in accordance with ASTM D2386 using K29790 Freezing Point Bath with Freezing Point Apparatus and Stirrer. The flash points were measured in accordance with ASTM D93 using automated Pensky-Martens flash point tester. The existent gum contents were measured in accordance with ASTM D381 using K33700 Existent Gum Evaporation Bath. The total acid numbers were determined in accordance with ASTM D664 through an acid-base titration using KOH as titrant. The copper corrosion grades were determined in accordance with ASTM D130 using Copper Strip Corrosion Test Bomb. The density and kinematic viscosity were determined in accordance with ASTM D4052 and ASTM D445, respectively.

3. Results and discussion

3.1. Comparison of the chemical compositions of aviation biofuels and aviation fuels

Table 3 and Figure 3 showed the relative contents of organic compounds in conventional aviation gasoline and aviation gasoline based on biomass. According to Figure 3, the aviation gasoline based on biomass was composed mainly of alkanes, aromatics, and olefins, with relative contents of 53.79 %, 16.52 %, and 9.29 %, respectively, compared to the values in aviation gasoline of 56.21 %, 28.29 %, and 10.38 %, respectively. The results indicated that the aromatics content in aviation gasoline based on biomass was 12.17 % lower than that of conventional aviation gasoline. The reduced aromatics were mainly Toluene.

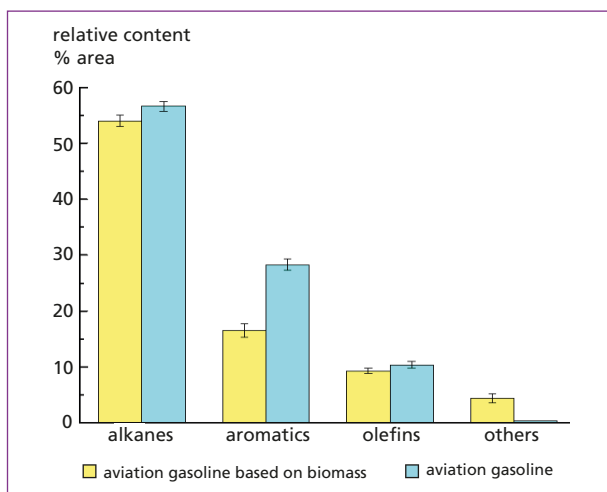


Figure 3:

Relative content of several compounds in aviation gasoline based on biomass and aviation gasoline

Table 3: Comparison of the chemical compositions of aviation gasoline based on biomass and conventional aviation gasoline

No.	compounds	relative content of conventional aviation gasoline	relative content of aviation gasoline based on biomass
		% area	
1	n-Butane	1.42	1.01
2	Ethanol	0	3.14
3	Isopentane	2.86	2.92
4	Pentane	0.93	0.97
5	Neohexane	1.64	1.33
6	Isohexane	5.51	5.07
7	Pentane, 3-methyl-	3.12	2.86
8	Hexane	2.24	2.17
9	2,4-Dimethylpentane	3.54	3.49
10	Benzene	3.93	3.4
11	2,3-Dimethyl-pentane	2.89	2.94
12	3-Methyl-hexane	1.29	0.67
13	2,2,4-Trimethylpentane	10.1	9.11
14	Heptane	0.95	0.94
15	2,2,3-Trimethyl-butane	5.97	6.05
16	2,3,4-Trimethyl-pentane	10.45	10.99
17	Toluene	10.52	0
18	2,3-Dimethylhexane	1.95	1.89
19	2,2,5-Trimethylhexane	1.35	1.38
20	1-Octane	0.23	0.39
21	Ethylbenzene	1.92	1.8
22	m-xylene	7.22	6.42
23	1,2-Xylene	3.14	2.9
24	Benzene,(1-methylethyl)-	10.38	9.29
25	n-Propylbenzene	0.29	0
26	1-Ethyl-3-methyl-benzene	0.74	0.77
27	1,2,3-Trimethyl-benzene	1.22	1.23
28	Tetraethyl-plumbane	0.39	0.49
29	1-Ethyl-4-hydroxybenzene	0	0.26
30	2,6-Di-tert-butyl-4-methylphenol	0	0.31
31	Palmitic acid	0	0.19

The aviation gasoline based on biomass also contained some phenols and organic acids. The relative content of phenols and organic acids in aviation gasoline based on biomass was 0.76 %, compared to 0 % in aviation gasoline. The organic acids were mainly composed of palmitic acid, with a relative content of 0.19 %, and the phenols mainly contained 1-ethyl-4-hydroxybenzene (0.26 %) and 2,6-di-tert-butyl-4-methylphenol (0.31 %). Although the bio-oil contained less 1-ethyl-4-hydroxybenzene and 2,6-di-tert-butyl-4-methylphenol than palmitic acid (Table 3), the aviation gasoline based on biomass had larger amounts of 1-ethyl-4-hydroxybenzene and 2,6-di-tert-butyl-4-methylphenol, illustrating that the capacity of these compounds to dissolve in aviation gasoline was higher than that of palmitic acid. In addition, 2,6-di-tert-butyl-4-methylphenol is an antioxidant, which can improve the anti-oxygenation of the aviation gasoline based on biomass.

Table 4: Comparison of the chemical compositions of aviation kerosene based on biomass and conventional aviation kerosene

No.	compounds	relative content of conventional aviation kerosene	relative content of aviation kerosene based on biomass
		% area	
1	Ethanol	0	0.67
2	1-Octane	0.54	0.52
3	Ethylcyclohexane	0.56	0.54
4	Octane, 2-methyl-	0.95	0.98
5	Octane, 3-methyl-	0.99	1
6	1-Ethyl-4-methylcyclohexane	0.8	0.8
7	n-Nonane	2.7	2.85
8	Propylcyclohexane	1.41	1.44
9	3-Methylnonane	0.94	0.98
10	2-Methyl-3-ethylheptane	0.93	0.95
11	Trans-hexahydroindan	0.75	0.74
12	Nonane, 2-methyl-	1.61	1.3
13	Nonane, 4-methyl-	1.4	1.47
14	3-Methylnonane	1.99	1.91
15	1,3,5-Trimethylbenzene	1.22	1.26
16	2-Heptene,3-ethyl-2-methyl-	1.10	1.10
17	1,1-Dimethyl cyclohexane	1.49	1.02
18	Decane	4.06	4.36
19	1,2,3-Trimethyl-benzene	0.8	0.76
20	2,4-Dimethylhexane	0.62	0.63
21	Nonane, 2,5-dimethyl-	0.95	0.98
22	Necane, 4-methyl-	0.92	0.96
23	n-Butylcyclohexane	1.42	1.48
24	Cyclopentane, pentyl-	0.85	0.89
25	Decane, 3-methyl-	1.07	1.25
26	Trans-perhydronaphthalene	1.42	1.5
27	Octane,5-ethyl-2-methyl-	0.84	0.85
28	5-Methyldecane	0.96	1.02
29	Decane, 2-methyl-	1.79	1.86
30	Decane, 3-methyl-	1.70	1.78
31	Citronellal	0.49	0.63
32	Undecane	4.38	4.88
33	Decane, 3,7-dimethyl-	1.29	1.33
34	Cyclohexane, pentyl-	0.99	1.04
35	Cyclododecane	1.09	1.12
36	Didecyl ether	0.49	0.54
37	Nonadecane	0	2.10
38	Undecane, 2-methyl-	1.57	1.65
39	Undecane, 3-methyl-	1.33	1.38
40	Naphthalene,decahydro-2,3-dimethyl-	0.95	0.96
41	1-Methyl-2-pentylcyclohexane	0.81	0.83
42	Dodecane	4.43	4.78
43	Tetratriacontane	0.82	0.82
44	Tridecane	2.64	2.75
45	Docosane, 1-bromo-	0.25	0
46	Cyclohexane, heptyl-	0.34	0
47	Tetradecane	0.62	0

As shown in Table 4 and Figure 4, the major chemical constituents of the aviation kerosene based on biomass and the conventional aviation kerosene were alkanes, with relative contents of 54.5 % and 52.05 %, respectively, a difference of 2.45 %, mainly resulting from nonadecane. Table 4 indicated that the alkanes of the bio-oil could dissolve in aviation kerosene, whereas the phenols and organic acids could dissolve in aviation gasoline. In addition, the relative contents and species of aromatics and olefins in the conventional aviation kerosene and aviation kerosene based on biomass were identical, which illustrates that the relative content and species of the aromatics and olefins in aviation kerosene did not change upon mixing the aviation kerosene with bio-oil.

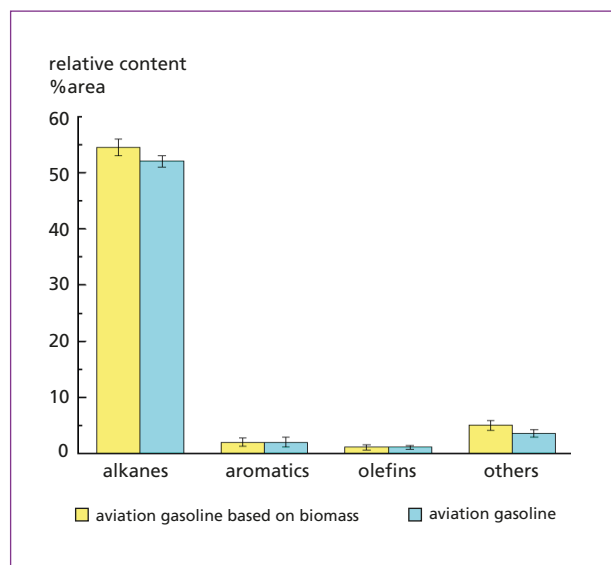


Figure 4:

Relative content of several compounds in aviation kerosene based on biomass and aviation kerosene

3.2. Physical and chemical properties of aviation biofuels

3.2.1. Total acid number and lower heating value of aviation biofuels

An increase in the total acid number in aviation fuels can erode the aircraft engine and reduce its working life [13]. In addition, the deposits from corrosion affect engine performance. Therefore, the total acid number of aviation biofuels must be analysed.

As shown in Figure 5, the total acid numbers of the aviation kerosene based on biomass and aviation gasoline based on biomass were 0.268 mg KOH/g and 0.504 mg KOH/g, whereas the total acid numbers of the conventional aviation kerosene and aviation gasoline were only 0.005 mg KOH/g and 0.34 mg KOH/g. The reason for the higher total acid number of the aviation gasoline based on biomass is that the acidic compounds (such as palmitic acid and phenols) of bio-oil dissolve in the aviation fuels during the mixing process. However, the total acid numbers of aviation gasoline based on biomass was still lower than the value (1 mg KOH/g) from specifications.

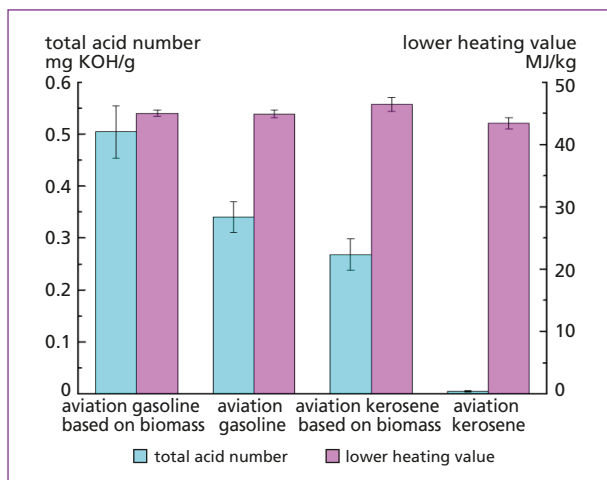


Figure 5:

Total acid number and lower heating value of aviation biofuels and aviation fuels

The aviation gasoline and aviation kerosene utilized in this study had the lower heating values of 44.9 MJ/kg and 43.44 MJ/kg, respectively. Comparing to aviation gasoline, the lower heating value of the aviation gasoline based on biomass increased by 0.2 % and exceeded the specifications, which was due to the dissolution of the oxygenated compounds (such as phenols and organic acids) from bio-oil into aviation gasoline [8]. Furthermore, the lower heating value of the aviation kerosene based on biomass increased by 7 % and the result could be associated with the increased content of alkanes (such as nonadecane) and oxygenated compounds (such as Citronellal and Didecyl ether) in aviation kerosene based on biomass. In addition, the lower heating values of the aviation biofuels also exhibited advantages compared to pure bio-oil. Xu [12], Wang et al. [11], and Hamieh et al. [4] provided values for the lower heating value of bio-oil from the liquefaction of biomass of 29.3 MJ/kg, 41.1 MJ/kg, and 40 MJ/kg, respectively, all of which were lower than the lower heating value of the aviation biofuels.

3.2.2. Freezing point and flash point of aviation biofuels

The freezing point and flash point are critical to the application of aviation fuels. As shown in Table 5, the freezing point of aviation gasoline based on biomass decreased compared to conventional aviation fuel from -58 to -70 °C, whereas the freezing point of aviation kerosene based on biomass decreased from -50.7 to -58 °C. Possible causes of these results were that aviation biofuels contained higher content ethanol with freezing point of -114 °C, which indicated the addition of alcohols could effectively decrease the freezing point. The flash point of aviation kerosene based on biomass was reduced by 19 °C because aviation kerosene based on biomass contained higher content Ethanol (0.67 %) and lower content Docosane, 1-bromo- (0), Cyclohexane, heptyl- (0) and Tetradecane (0), which leads to its higher volatility at high temperatures. However, comparing to aviation gasoline, the flash point of aviation gasoline based on biomass was higher. Possible causes of which were that the content of low flash point compound (n-Butane, Toluene) decreased and the content of high flash point compound (1-Ethyl-4-hydroxybenzene, 2,6-Di-tert-butyl-4-methylphenol, Palmitic acid, ethanol) increased in aviation gasoline based on biomass.

Table 5: Physical and chemical properties of aviation biofuels and aviation fuels

Properties	Unit	Aviation kerosene	Aviation kerosene based on biomass	Aviation gasoline	Aviation gasoline based on biomass	Bio-oil
Freezing point	°C	-50.7	-58	< -58	< -70	-8
Flash point		42	23	< 10	5	50
Existent gum	mg/100 ml	1.7	105	1.6	198	300
Copper corrosion 3 h at 100 °C		1	1b	1	1b	3a
Density at 20 °C	kg/m ³	820	840	748	800	1,100
Kinematic viscosity at 20 °C	mm ² /s	1.45	1.512	Nr	Nr	5.81
Water content	wt %	-	0.36	-	0.32	10.16
Octane rating	MON	Nr	Nr	95.5	98.7	Nr

Nr: Not required

-: None

3.2.3. Density and kinematic viscosity of aviation biofuels

Density is an important parameter for calculating the volumetric output from pumps and injectors, however, kinematic viscosity has a significant effect on spray characteristics of engine. As shown in Table 5, aviation biofuels had higher density than aviation fuels, which may be due to the presence of water in aviation biofuels [2]. Comparing to aviation kerosene, the kinematic viscosity of aviation kerosene based on biomass was increased by 0.062 mm²/s because of the increasing relative content of longer chain length compound [6], such as nonadecane (2.1 %).

3.2.4. Octane rating and water content of aviation biofuels

The octane rating is a key parameter for describing the anti-detonating quality of aviation gasoline, which was measured as ASTM D2699. The results were shown in Table 5 and it was found that the octane rating of aviation gasoline based on biomass was higher than that of aviation gasoline due to the higher content of Tetraethyl-plumbane (0.49 %). In addition, Table 5 showed the water content (measured as ASTM D4928-12) of aviation biofuels was higher, which is related to the high water content (10.16 wt-%) of bio-oil from liquefaction with water as solvent.

3.2.5. Existent gum contents and copper corrosion grades of aviation biofuels

Table 5 showed the existent gum contents and copper corrosion grades of aviation biofuels and conventional aviation fuels; the aviation kerosene based on biomass and aviation gasoline based on biomass contained 62- and 123-fold more existent gum, respectively. Most of the existent gum was from the bio-oil. The copper corrosion grades of the aviation biofuels and conventional aviation fuels were 1 and 1b, respectively, indicating that these fuels cause no damage to the copper of the mechanical equipment.

4. Conclusion

The results revealed the compatibility of bio-oil obtained from hydrothermal liquefaction based on chemical-biological pretreatment with aviation fuels. Bio-oil was compatible with aviation fuels with net heating values, freezing points and copper corrosion, as well as kinematic viscosity. However, the density, water content, flash point and existent gum of aviation biofuels were out of specifications. In result, the bio-oil would have better compatibility with aviation fuels after refining, such as catalytic cracking, dehydrating and degumming. In addition, the total acid number of aviation kerosene based on biomass was found to be higher than 0.015 mg KOH/g, the maximum value specified from standard, which indicated the bio-oil should be addressed by esterification or catalytic hydrogenation. The total acid number of aviation gasoline based on biomass was below the specification maximum (1 mg KOH/g) and the octane rating of that was higher than standard value.

Bio-oil from hydrothermal liquefaction based on chemical-biological pretreatment could be more perfectly compatible with aviation gasoline only requiring further dehydrating and degumming.

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