

Soot Formation of Aviation Bio-Fuels in Diffusion Flames

THONG DUC HONG¹, OSAMU FUJITA², TATANG H. SOERAWIDJAJA³

¹Department of Automotive Engineering, Ho Chi Minh City University of Technology, VNU-HCM, Vietnam

²Division of Mechanical and Space Engineering, Hokkaido University, Sapporo, Japan

³Department of Chemical Engineering, Institut Teknologi Bandung, Bandung, Indonesia
Email: hongducthong@hcmut.edu.vn, ducthonghong@gmail.com

Abstract: Co-annular smoke-free laminar diffusion wick-fed flames of aviation bio-paraffins and their blended with various amounts of propylbenzene of 10, 20, 25 Vol.% have been used to study soot formation. A light extinction method is adopted to determine the total soot volume (TSV) as a function of flame height (FH) and fuel mass consumption rate (FMCR). The empirical models have been built to predict aviation bio-paraffins/propylbenzene (Bio-P/PB) mixtures as the function of two variables of FMCR and concentration of propylbenzene content (%PB). TSVs of Bio-P/PB mixtures increase with increasing flame height, FMCR and %PB. The effect of flame height, FMCR and concentration of propylbenzene content on soot formation are respectively expressed as the quadratic, power law and linear functions. The results of current work create a database for optimizing the trade-off impacts of aromatic in aviation biofuel. This information is of high importance when blending aromatic to aviation bio-paraffins, which is produced from triglycerides and fatty acids in the vegetable by hydrotreating process, for using as a fuel in aircraft engines.

Keywords: Aviation, Bio-paraffins, Propylbenzene, Soot Formation, Wick-Fed, Diffusion Flame

Introduction:

Alternative fuels are known as a feasible solution for the challenges of rapid exhaustion of fossil-based liquid fuels and climate change. In practically, there are a number of achievements in researches and successful applications of biofuels on stationary and automobile engines. The renewable fuels have received more attention in the airline industry [1 – 17] since 2008 when European Commission approved the European Union Emissions Trading Scheme (EU ETS) to include the civil aviation sector. Directive 2008/101/EC of the European Parliament and Council [18] agrees that from 2012, CO₂ emissions from all commercial flights to, from, and within the European Union are covered by the EU ETS. Under the EU ETS, biofuels are considered CO₂ neutral [19, 20] and the airline can benefit from an exemption from the need to surrender allowances and credits.

There are currently two main production processes for alternative aviation fuels as following: hydroprocessed renewable jet and Fischer-Tropsch jet. These both processes produce the final products are bio-paraffin compounds, in which the carbon chain lengths fall within the boiling range of the jet fuel (number of carbon C₈ – C₁₆). In order to satisfy the freezing point, lower heating value, density requirements of aviation fuel standards, these bio-jet paraffins are then to be blended with appropriate proportion of aromatics (<25% by volume) to the so-called “drop in” jet fuel. Blends of “drop in” jet biofuel with fossil-based aviation kerosene at various volumetric proportions could be used to power jet aircrafts. Aromatic is added to aviation bio-paraffins with a view to (a) prevent fuel leakage in the aircraft fuel system and tanker trucks [7, 17] as a result of increasing seal swell; and (b) improve the freezing points [21].

However, aromatic is also known as a substance which has strong effect on soot formation. Soot particles can cause overheat in combustion chamber, reducing the efficiency and long life of engines [22, 23]. In addition, when soot particles exit from the engine, they cause serious health effects and global warming [24 – 28]. Therefore, the soot characteristics of aviation biofuels need to be addressed before proceeding with other tests on aircraft engines to apply a new fuel to aviation use.

The experiments reported here investigated soot emissions in a smoke-free diffusion wick-fed flames established over a laminar co-flow burner of aviation biofuel, which had represented in our previous study [21], and its blends with propylbenzene of 10, 20 and 25 Vol.%. We measured the total soot volume (V_S) and fuel mass consumption rate (\dot{m}_f) along with the visible flame height (H_f), $\dot{m}_f = f(H_f)$ and $V_S = f(H_f)$, of each flame by using light extinction method. The relationship between total soot volume and fuel mass consumption rate, $V_S = f(\dot{m}_f)$, was then calculated. Total soot volume, $V_S = f(\dot{m}_f, \%PB)$, was also built as a function of the two variables of fuel mass consumption rate, \dot{m}_f , and propylbenzene percentage content, %PB, for aforementioned biofuel/propylbenzene mixtures.

The objective of this work is to assess the effect of each individual factor such as flame height, fuel mass consumption rate, and concentration of propylbenzene on the soot propensities in diffusion flames of the aviation biofuel. This is the important database for optimizing the trade-off impacts of aromatic in aviation biofuel and thus the optimal blending ratio of propylbenzene in biofuel can be predicted for the purpose of practical application.

Experiment description:

Materials

Propylbenzene and aviation biofuel were the tested fuels. Propylbenzene (C₉H₁₂) was supplied from the Merck Millipore Company, Japan.

The aviation biofuel was supplied by the Research & Development Division of the Pertamina Oil Company, Indonesia. The biofuel was manufactured following the production process previously described in the literature [21]. The process based on the hydrotreating method without cracking and isomerization steps and coconut oil was used as the feedstock for this process. The compositions of the aviation biofuel are the paraffinic compounds containing predominantly undecane (n-C₁₁H₂₄) and dodecane (n-C₁₂H₂₆) and thus the aviation biofuel is called as bio-paraffins.

The chemical reactions of lauric oil and triglyceride, which are the dominant components in coconut oil, to form undecane and dodecane are illustrated in Fig. 1 and the common properties of bio-paraffins are reported in Table 1.

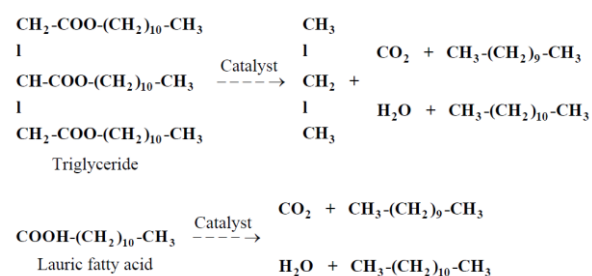


Figure 1: Molecular transformation steps of bio-paraffins production process [21].

Table 1: The common properties of aviation bio-paraffins [21].

Property	ASTM D1655 [29]	Bio-paraffins
Distillation temperature:		
Initial BP, °C	Report	141
10% rec, °C	Max. 205	191
50% rec, °C	Report	218
90% rec, °C	Report	283
Final BP, °C	Max. 300	308
Flash point, °C	Min. 38	47
Density at 15°C, kg/m ³	775 – 840	759
Freezing Point, °C	Max. -47.0	9.5
Viscosity at -20°C, mm ² /s	Max. 8.0	6.485
Lower heating value, MJ/kg	Min. 42.8	42.48
Sulfur, total, wt%	Max. 0.3	11 ppm

The four fuels considered in this study are listed in Table 2. Fuel (1) to fuel (4) respectively represent the mixtures of bio-paraffins and propylbenzene of 0, 10, 20 and 25 Vol.%.

Table 2: The samples of experimental fuel.

Fuels	Sign	Components, Vol.%	
		Bio-paraffins	Propylbenzene
(1)	Bio-P100%	100	0
(2)	Bio-P90%+10%PB	90	10
(3)	Bio-P80%+20%PB	80	20
(4)	Bio-P75%+25%PB	75	25

2.2 Experimental procedures

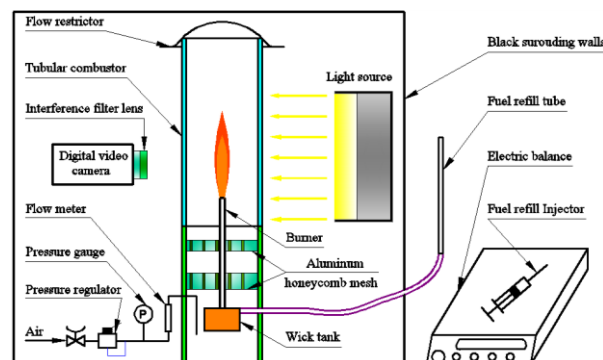


Figure 2: The schematic diagram of the experimental setup.

Figure 2 shows the schematic diagram of the experimental setup for determining soot formation. Soot characteristics, which are generated from co-annular smoke-free laminar diffusion wick-fed flames, were obtained by light extinction measurements in the current work. The flame height (FH) was controlled by adjusting the height of the wick extended above the wick tube rim which had the inner and outer diameters of 7 and 8mm, respectively. The tubular combustor was made of Pyrex glass with the diameter of 90 mm and length of 250 mm.

The combustion air was supplied from the bottom of the tubular combustor at the constant flow rate of 30 lit/minute (mean air velocity of 7.86 cm/s) for all experiments. To obtain the uniform flow field in the combustion zone, the air was supplied through a double aluminum honeycomb structure of 750 cells per square inch. A flow restrictor was placed at the exhaust of the tubular combustor to achieve a highly stable flame by reducing any air recirculation. The flow restrictor consisted of a 120 mm diameter stainless steel bowl with a center hole and three concentric rings of 6 mm diameter holes. The experimental set-up was enclosed in black colored surroundings to reduce extraneous impacts from the environment.

An XBPA540 interference filter lens, which was purchased from the Asahi Spectra Company, UAS, was used to select the wave length for the extinction image. This filter lens only allows optical access of wave length of 540 nm. A Japanese Panasonic HDC-TM750 digital camera, in which the filter lens was

mounted on, was used to record the direction and shadow of the back light. These records were converted to the still images. Then the stable burning flame images and the extinction images were analyzed to compare their light intensity by a Matlab program. The total soot volumes (TSVs) were estimated by applying the Beer – Lambert law and Mie theory in the Rayleigh limiting to the attenuated backlit image.

Fuel mass consumption rates (FMCRs) were measured by using a Shimadzu UX2200H digital balance with readability of 0.01g, a fuel-refilling injector and a timer. For enhanced accuracy, each measurement was performed with a fuel mass that is larger than 1g and each test point was repeated thrice to yield the average reading. The repeatability was matched over 95% for each test. This means that the FMCR measurement can be reliably analysed from the experimental data.

TSV and FMCR were determined as the functions of FH for the 4 experimental fuels in the limitation of their smoke-free flames. Then, the correlation between TSV and FMCR was calculated. All experiments were done at the room temperature and atmosphere pressure.

Soot particle measurement by extinction of light

The Beer – Lambert law states that there is a logarithmic dependence between the transmission of light through a substance and the extinction cross section, C_{ext} , the number density, N , of the particles of the absorber and the distance light travels through the substance (the path length), L .

$$\frac{I}{I_0} = e^{-LNC_{ext}} \quad (1)$$

$$C_{ext} = A_C Q_{ext} = \frac{\pi d^2}{4} Q_{ext} \quad (2)$$

Where: I , I_0 are respectively transmitted and incident light intensity, A_C is the cross section area of the particle, d is the diameter of the particle, and Q_{ext} is the extinction efficiency.

According to Mie scattering theory for sphere [30, 31], when the particle size is much smaller than the wavelength of the incident radiation ($x \ll 1$) and if $|m|x \ll 1$, the extinction efficiency is approximately:

$$Q_{ext} = 4x \operatorname{Im} \left\{ \frac{m^2 - 1}{m^2 + 2} \right\} \quad (3)$$

Where: $x = \pi d / \lambda$ is the particle size diameter, λ is the wavelength of the incident light, $m = n - ik$ is the complex refractive index of the particle, n and k are respectively the real and imaginary part of m , and Im is the imaginary value of a complex number.

Substituting (2) and (3) to (1) we have:

$$\ln \left(\frac{I}{I_0} \right) = - \frac{\pi^2 d^3 NL}{\lambda} \operatorname{Im} \left\{ \frac{m^2 - 1}{m^2 + 2} \right\} \quad (4)$$

The soot volume fraction can be expressed as a function of the total number of primary particles per unit volume, N , and the soot particle diameter, d , as follows:

$$f_v = \frac{\pi}{6} d^3 N \quad (5)$$

Substituting (5) to (4) we have:

$$f_v = - \frac{\lambda \ln \left(\frac{I}{I_0} \right)}{6\pi L \operatorname{Im} \left\{ \frac{m^2 - 1}{m^2 + 2} \right\}} \quad (6)$$

It is assumed that all soot particles are the same size, soot volume fraction based on optical measurements can be calculated from equation (6) by obtaining the intensity of the transmitted light and incident light as well as determining the path length of the extinction and the selected wavelength of the incident light.

In this work, $m = 1.57 - 0.51i$, which was used for analyzing soot, was computed from the empirical data of Dalzell and Sarofim [32], who concluded that the optical properties of different carbons depend to a small extent on the differences in the molecular structure and to a larger extent on the differences in H/C ratio. As mentioned earlier, the biofuel expected is a paraffinic compound and has similar average chemical formula with dodecane so that it is valid for using the same refractive index to analyze soot in this study.

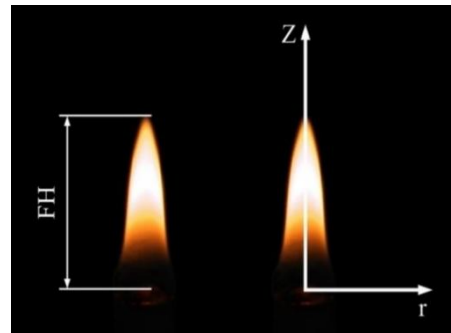


Figure 3: The total soot volume was measured within the entire volume of visible flames.

The attenuation measurement of backlight is a kind of in-line method where the corresponding field distribution function, $f_v(Z, r)$, was obtained using a three-point Abel deconvolution algorithm [33]. Based on the measured soot volume fraction distributions, the total volume of the soot particles, V_s , within the entire visible flame volume can be calculated using the following expression:

$$V_s = \int_0^{FH} dZ \int_0^R f_v(Z, r) 2\pi r dr \quad (7)$$

Where: R is the radius of the luminous flame envelope at height Z , and FH is the visible flame height (the height of the luminous zone). The calculation of V_s was performed using the measured

soot volume fraction distributions up to $Z = FH$ (see Fig. 3). Above that height, only negligible amounts of soot existed.

Results and discussions:

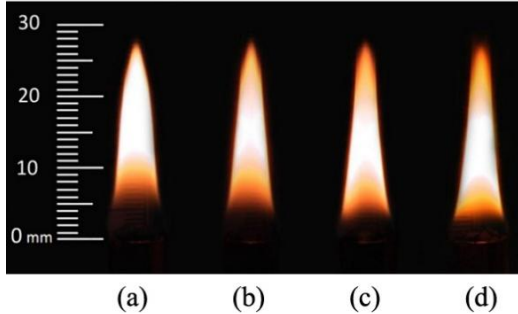


Figure 4: Image of flames at the height of 28mm, (a) – (d) corresponding with fuel samples (1) – (4), respectively.

The flame images of 4 different fuel samples are provided in Fig. 4. The flames were recorded at the height of 28 mm. The shapes and appearances of the flames of bio-paraffins/propylbenzene (bio-P/PB) mixtures changed significantly with the concentration of aromatic.

Increasing propylbenzene concentration caused the flame diameters to narrow and the flame fronts changed from convex surface to concave surface. The invisible sooting regions existed in the lower portion of the flames diminished and the yellow luminous zones in the upper portion of the flames moved upstream (towards burner tip), suggesting that soot inception were formed near the burner exit.

The more the propylbenzene concentration, the higher luminosity of flames was obtained. Pure bio-paraffins had the closed tips. When aromatic was added to bio-paraffins, the tips of flames widened with the appearance of sooty flame “wings”. It had been predicted that soot loading grew when increasing propylbenzene content. The physical appearances of the flame structures observed in the current study have similar tendency with the work of Kobayashi et al. [34] studied on a laminar diffusion pool combustion flames of benzene (C_6H_6) and hexane (C_6H_{14}).

Figure 5 shows the back light images of the same fuel at different flame heights (Figs. 5a and 5b) and different fuels at the same flame height (Figs. 5b and 5c). The shadows of the lights caused by the presences of soot are clearer at the higher flame and higher concentration of propylbenzene in the mixture. This suggests that soot loading increases with the flame height and the proportion of propylbenzene in the mixture.

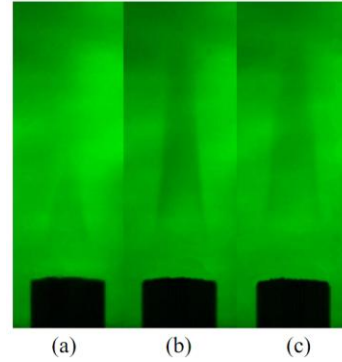


Figure 5: Images of back light – (a): fuel (4) exhibiting a flame height of 14 mm, (b): fuel (4) exhibiting a flame height of 28 mm, (c): fuel (2) exhibiting a flame height of 28 mm.

Figure 6 presents FMCR as a function of the FH for Bio-P/PB mixtures. There are closely linear FH to FMCR relationships of pure bio-paraffins. This result is consistent with the theory [35, 36] and the previous experimental studies of Olson et al. [37] and Glassman et al. [38].

According to the diffusion flame theory [36, 37], the flame height, H_f , of a cylindrical diffusion flame can be described by:

$$H_f = \frac{22400 \dot{m}_f}{4\pi \cdot D_0 \cdot MW} \left(\frac{T_0}{T_f} \right)^{0.67} \frac{1}{\ln(1+S^{-1})} \quad (8)$$

Where: \dot{m}_f is the fuel mass consumption rate, D_0 is the diffusion coefficient at 300K, MW is the molecular weight, T_f and T_0 are respectively the mean flame temperature and initial fuel/air temperature, and S is the volume of air required to burn a volume of fuel.

Employing the following relations:

$$\ln(1+S^{-1}) \sim S^{-1} \quad (9)$$

We find that:

$$H_f = \frac{22400 \dot{m}_f}{4\pi \cdot D_0 \cdot MW} \left(\frac{T_0}{T_f} \right)^{0.67} S \quad (10)$$

Equation (10) implies that the FH is expected to directly scale with the FMCR if the other parameters keep constant. The data in this work, however, indicates the relationship became non-linear when adding propylbenzene to bio-paraffins. The higher the propylbenzene content the lower the slope of the curve was obtained, and the slope turned non-linear, that is, decreasing in slope with an increase in the FH. In other words, the flame height, which contained propylbenzene, was higher than what expressed in Eq. (10). The higher FH in this case was a consequence of the increase in the radiation heat loss associated with the higher soot formation that lead to the reduction of the mean flame temperature. Another reason is that the soot increased in the flame might have changed the flame length determination mechanism behind the Eq. (10), e.g. the necessity to consider soot oxidation reaction time rather than

oxygen diffusion time toward the centre of the flame. The results of the present work fairly agreed also with Roper et al. [39] in which an anomaly (nonlinear relationship was found between the visible flame height and fuel consumption) occurred for propylene

with the authors' explanation that when soot concentrations are high, a soot oxidation zone follows the diffusion flame, causing a large increase in the visible flame size.

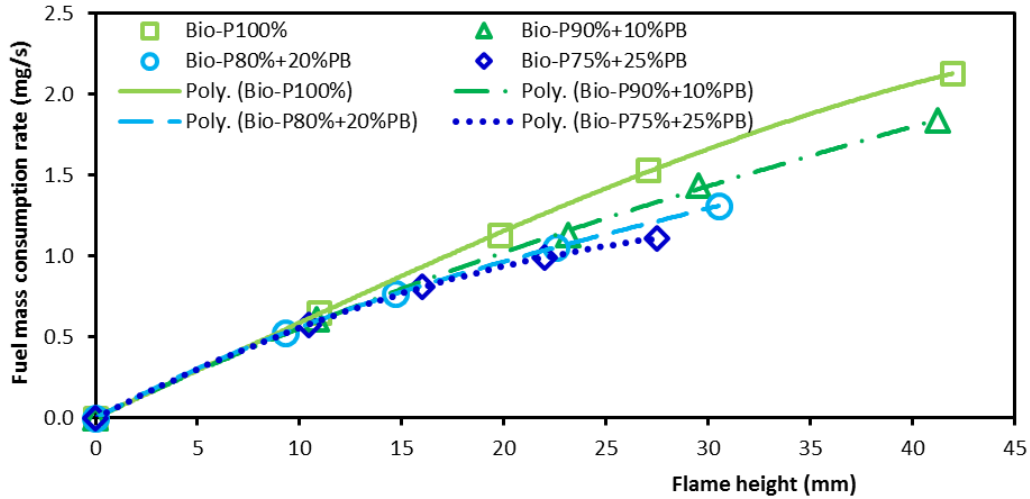


Figure 6: Relation between FMCR and FH of 4 fuel samples.

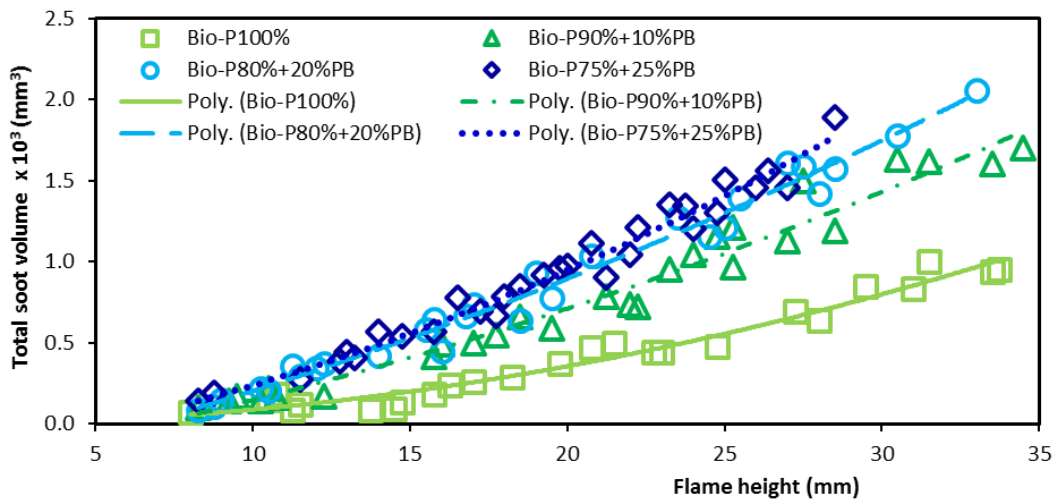


Figure 7: TSV versus FH of Bio-P/PB mixtures.

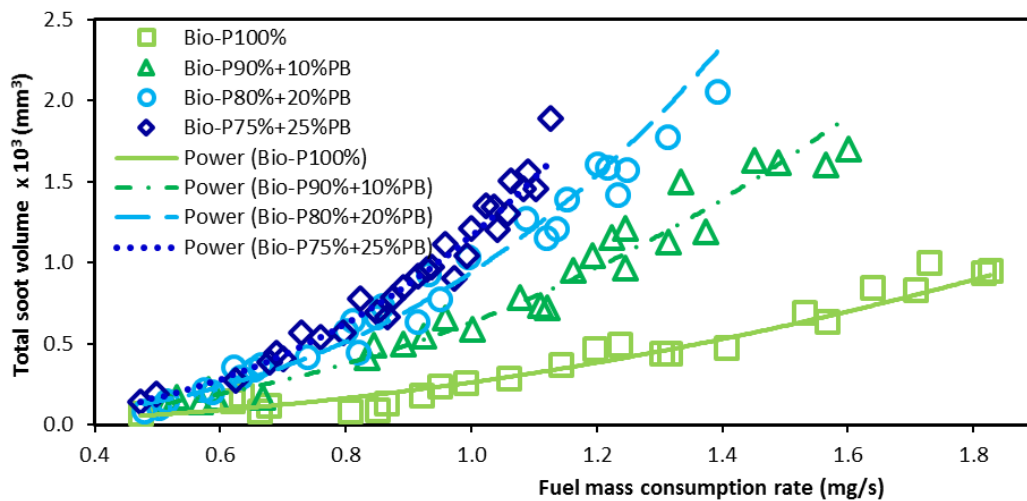


Figure 8: TSV versus FMCR of Bio-P/PB mixtures.

Table 3: The functions of TSV versus FH and FMCR for 4 different samples of tested fuels.

Fuel	TSV, V_s , and coefficient of determination, R^2	
	Versus FH, H_f	Versus FMCR, \dot{m}_f
Bio-P100%	$V_s = 0.0009(H_f)^2 + 0.0003H_f - 0.0045$ $R^2 = 0.9626$	$V_s = 0.2609(\dot{m}_f)^{2.0947}$ $R^2 = 0.8881$
Bio-P90% + 10%PB	$V_s = 0.0008(H_f)^2 + 0.0333H_f - 0.2648$ $R^2 = 0.9657$	$V_s = 0.6312(\dot{m}_f)^{2.3522}$ $R^2 = 0.9780$
Bio-P80% + 20%PB	$V_s = 0.0008(H_f)^2 + 0.0450H_f - 0.3273$ $R^2 = 0.9819$	$V_s = 0.9388(\dot{m}_f)^{2.7129}$ $R^2 = 0.9678$
Bio-P75% + 25%PB	$V_s = 0.0014(H_f)^2 + 0.0294H_f - 0.1950$ $R^2 = 0.9714$	$V_s = 1.1648(\dot{m}_f)^{2.7963}$ $R^2 = 0.9806$

Figures 7 and 8 reveal soot formation of Bio-P/PB mixtures along with FH and FMCR, respectively. Soot loading increases with FH, FMCR and the proportion of propylbenzene content (%PB). The increments of soot versus FMCR are stronger than that versus FH. The lines represent the best fit curves which are the quadratic functions for TSV, V_s , – flame height, H_f , relationships and the power law forms for TSV, V_s , – FMCR, \dot{m}_f , relationships. The equations and their coefficients of determination are shown in Table 3.

From the experimental data obtained above, the authors attempted to develop empirical models that were able to predict the TSV, $V_s(\dot{m}_f, \%PB)$ as a function of two variables of FMCR, \dot{m}_f , and the percentage of propylbenzene content, %PB, for Bio-P/PB mixtures with the following steps together with the accompanying assumptions as:

- i. The effects of \dot{m}_f and %PB are independent on TSV, so the function of $V_s(\dot{m}_f, \%PB)$ has the form:

$$V_s(\dot{m}_f, \%PB) = f(\dot{m}_f) \times f(\%PB) \quad (11)$$

Where $f(\dot{m}_f)$, $f(\%PB)$ are respectively the individual functions of the effect of FMCR and %PB on TSV.

- ii. The function of the effect of FMCR on TSV, $f(\dot{m}_f)$, is assumed to be equal to the function of TSV for pure bio-paraffins, $V_s(\dot{m}_f, 0)$.

From Table 3 we have:

$$f(\dot{m}_f) = V_s(\dot{m}_f, 0) = V_{s, \text{bio-paraffins}} = 0.2609(\dot{m}_f)^{2.0947} \quad (12)$$

- iii. The differences of TSV between pure bio-paraffins and its mixtures with propylbenzene of 10, 20, 25 vol.% are caused by the effect of propylbenzene, including the synergistic effects of fuel compounds, the differences of fuel properties such as density, viscosity, the net heat of combustion, the boiling point etc. From Eqs. (11) and (12) we have the function of effect of %PB on TSV for Bio-P/PB mixtures:

$$f(\%PB) = \frac{V_s(\dot{m}_f, \%PB)}{0.2609(\dot{m}_f)^{2.0947}} \quad (13)$$

By substituting all the experimental data to the equations (13), we can find the best fit functions of the effect of %PB on TSV, $f(\%PB)$, for Bio-P/PB mixtures.

The total coefficient of determination, R_{total}^2 , is calculated as follow:

$$R_{\text{total}}^2 = 1 - \frac{\sum_i^n (y_i - f_i)^2}{\sum_i^n (y_i - \bar{y})^2} \quad (14)$$

Where: n is the number of observations, y_i and f_i are the observed and predicted values respectively, and \bar{y} is the mean of the observed data.

The function of TSV for Bio-P/PB mixtures:

$$V_s(\dot{m}_f, \%PB) = 0.2609(\dot{m}_f)^{2.0947} * [11.828(\%PB) + 1.1061] \quad (15)$$

$$R_{\text{total}}^2 = 0.93$$

Equations (15) show that TSV is linearly proportional to the concentration of propylbenzene. With a given FMCR, TSVs of Bio-P/PB mixtures increase respectively about 10.7% for each 1% Vol. propylbenzene added to the mixture. It is easy to see the significant effect of aromatic on soot formation of the mixture.

Conclusions:

Aviation bio-paraffins/propylbenzene (Bio-P/PB) mixtures have been investigated on soot formations in co-annular wick-fed laminar diffusion flames and light extinction method was used to determine their total soot volumes (TSVs).

The work results in the conclusions as follows:

There are closely linear relationships between flame height (FH) and fuel mass consumption rate (FMCR) with pure bio-paraffins. However, the relationship becomes nonlinear when adding propylbenzene to bio-paraffins due to the significant increase of soot formation in flame. The higher propylbenzene content leads to the lower and non-linear slope of the curve.

TSVs of Bio-P/PB mixtures increase with the increasing FH, FMCR and the proportion of propylbenzene content (%PB). The increment trends of TSVs are non-linear to FMCR and FH. The trends versus FMCR and FH are represented by a power law form and a quadratic function, respectively.

The empirical models were built to predict the TSVs of Bio-P/PB mixtures as a function of two variables which are FMCR and %PB. From the models, TSV is linearly proportional to %PB. With a given FMCR, TSVs of Bio-P/PB mixtures increase about 10.7% for each 1% propylbenzene added to the mixture. It implies the significant effect of aromatic on soot formation.

This study has an important significance when considering optimal blending ratio of aromatics and bio-paraffins to utilize the advantages of aromatics for achieving the stringent requirements of aviation fuel as well as to limit the harm which they cause to the engines, people and environment. Although the combustion in the combustion chamber of gas turbine engine is quite different from what occurred in this experimental condition, the present work supplies a valuable fundamental background to predict the complex mechanisms and provides useful database for further experiments in a real engine.

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