Hydraulic and Thermal Effects of Coke Deposition during Pyrolysis of Hydrocarbon Fuel in a Mini-Channel

ARTICLE *in* ENERGY & FUELS · JUNE 2012

Impact Factor: 2.79 · DOI: 10.1021/ef3008079

7 AUTHORS, INCLUDING:

Zhaohui Liu
Xi'an Jiaotong University
20 PUBLICATIONS **58** CITATIONS
SEE PROFILE

Jianguo Yan
Xi'an Jiaotong University
12 PUBLICATIONS **36** CITATIONS
SEE PROFILE

energy&fuels

Article

pubs.acs.org/EF

Hydraulic and Thermal Effects of Coke Deposition during Pyrolysis of Hydrocarbon Fuel in a Mini-Channel

Zhao H. Liu,[†] Qin C. Bi,*[,†] Yong Guo,[†] Xue S. Ma,[‡] Zhu Q. Yang,[†] Jian G. Yan,[†] and Shen L. Hu[‡]

[†]State Key Laboratory of Multiphase Flow in Power Engineering, Xi'an Jiaotong University, Xi'an 710049, China

[‡]Science and Technology on Scramjet Laboratory, The 31st Research Institute of CASIC, Beijing 100074, China

ABSTRACT: Coke deposition in a mini-channel reduced the effective diameter greatly during pyrolysis of hydrocarbon fuel. In the paper, the hydraulic resistance method (HRM) was introduced to quantitatively evaluate the coke deposition. Pyrolysis of hydrocarbon fuel was carried out at fuel temperature up to 725 °C at supercritical pressure of 4 MPa in an electrically heated mini-channel (heated length, 508.0 mm; inner diameter, 2.0 mm; wall thickness, 0.5 mm). The test run was maintained at steady state for 30 min. The post-test coke evaluation by HRM is nondestructive and onboard performed at normal temperature and pressure. Compared with the optical microscope visualization, the HRM was validated to be feasible and effective. Experimental results show that the thickness of coke deposit along the channel, varying from several to about 130 μm, is a function of film temperature (average of the wall and bulk fuel temperature). Contributing to the coke deposition during fuel pyrolysis, the hydraulic resistance continuously increased. However, depending on the deposit thickness, the thermal effects can be contrary. The gully coke nature at high *Re* turbulent flow and the large deposit thickness as a thermal resistance can dominate to enhance and reduce the heat transfer, respectively.

1. INTRODUCTION

Endothermal hydrocarbon fuel is used as both propellant and coolant in present military aircraft. Regenerative cooling technology is the most promising solution to discharge the huge amount of redundant heat load.[1] At a Mach-8 scramjet, the heat sink capacity of hydrocarbon fuel is required to be above 3.5MJ/kg. Therefore, the fuel temperature exceeds 750 °C after the cooling passages (about 1.0 mm equivalent diameter). Development of endothermal hydrocarbon fuel with high heat sink is profoundly limited by its thermal instability. The key challenge for hydrocarbon fuels is the difficulty of improving the cooling capability of the fuel but without having the vehicle's life limited by carbonaceous deposit in the passages.[2]

Exposure of fuels to high temperatures results in the coke formation. To pursue a hydrocarbon fuel with good coking property, how to define and evaluate the coking characteristic is the first and key problem. We are concerned with "when" and "how much" deposit will occur under the real fuel conditions.[3] Carbonaceous products can dissolve in the fuel or in a more dangerous way deposit on the internal surface of reactor. Therefore, it is necessary to identify and measure the coke layer formation.

Generally, there are mainly two types of tests: flowing and static.[4] The reported thermal stability test rigs and conditions were summarized by Heneghan.[5] In the flowing tests, hydrocarbon fuel flowed through a heated tube. At the end of test, the tube was cut into sections, and carbon burnoff or gravimetric analysis of the tube was performed. Carbon burnoff used to evaluate the coke quantity is a popular method and always finds its application in the past and present. The carbon deposit on each section was measured using a LECO RC-412 Multiphase Carbon Determinator.[6−8] The method is based on temperature-controlled heating of the sample in oxygen, with continuous recording of the amount of carbon oxides evolved, as a function of temperature.[9] The carbon burnoff and the preparation before test are a series of complicated procedures and are performed during downtime with destructive measures. In a static-type test, the quartz crystal microbalance (QCM)[4] was used to make in situ measurement of deposition at low temperatures. The QCM device can be maintained at a pressure of 1000 psig and fuel temperature up to 300 °C. The destructive measure and limited temperature are the shortcomings of the above two methods. Is there any method without the disadvantages?

Coke deposition increases the hydraulic resistance of the test channel. The coke measure by considering the hydraulic effect can be realized at the original test channel without the fuel temperature being limited. Deposit of heavy organics always occurred in petroleum production,[3,10] transportation,[8,9] and the processing industry.[11,12] Pressure drop was always employed to analyze the fouling conditions in the transportation pipelines and heat exchangers. The hydraulic effect was also introduced to find the deposit velocity in an ice slurry fluid flow.[13,14]

The hydraulic effect is used as a witness of coke formation in test channel.[15] The mass flow loss at a given pressure drop was measured to quantify the amount of coke that occurred throughout the test sections.[16] However, to our knowledge, little literature had reported that the hydraulic resistance was applied to calculate the hydraulic diameter of test channel and to evaluate the amount of coke deposition.

In this paper, a convenient and efficient method is introduced to find the amount of coke deposition after the pyrolysis of hydrocarbon fuel. The method is based on the

Received: May 10, 2012
Revised: May 29, 2012
Published: May 30, 2012

dx.doi.org/10.1021/ef3008079 | *Energy Fuels* 2012, 26, 3672−3679

change of hydraulic resistance. No additional operation is needed to be performed for the flow channel, but there is a need to measure the hydraulic resistance of test tube at normal temperature and pressure before and after the coking test. It is significant that the method can be applied without the structure of test channel being limited, which may be a single straight channel, a serpent channel in a flat plate heat exchanger, or the channels in a whole scramjet.

Results of the above hydraulic resistance method (HRM) were validated by the visualization method (VM) to identify the feasibility and accuracy. After the pressure drop test, the channel was cut into sections, and the coke layer was observed with an optical microscope (OM) and scanning electron microscope (SEM) visualization. In the last part of the paper, thermal and hydraulic effects of coke deposition are analyzed.

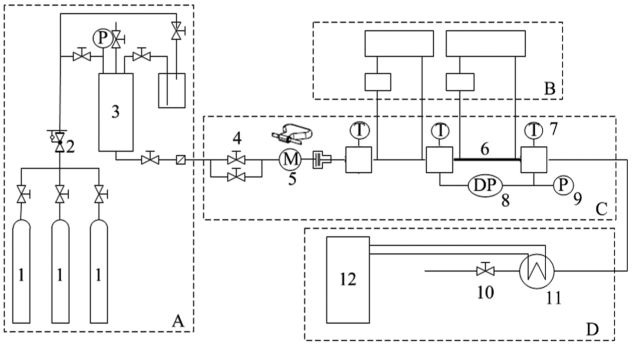


Figure 1. Schematic figure of the experimental system. (1) Nitrogen cylinder; (2) Pressure reducing valve; (3) Fuel tank; (4) Flow control valves; (5) Mass flow meter; (6) Test channel; (7) K-type sheathed thermocouple; (8) Differential pressure transducer; (9) Pressure transducer; (10) Back pressure valve; (11) Heat exchanger; (12) Water chiller.

2. EXPERIMENTAL SECTION

Experiments were conducted to study coking properties of hydrocarbon fuel. Figure 1 shows the simplified experimental system. It contains five sections: the fuel feed system, A; electrical heating system, B; test channel and measurement system, C; cooling system, D; and data acquisition system.

Hydrocarbon fuel is fed by high pressure nitrogen, 1, and then passes through the electrically heated test channel 6 from high pressure feed tank 3. Valves 4 before the test channel are used to precisely control the mass flow rate. Outlet pressure is controlled by the back pressure valve 10 after the fuel cooled. A preheated channel is set before the test channel to regulate the fuel inlet temperature.

The mass flow rate of hydrocarbon fuel was measured before the inlet of test section by a Micro Motion Coriolis mass flow meter. The pressure was measured at the outlet by Rosemount pressure transducer. Pressure drop of test channel was measured by Rosemount differential pressure transducer. All the measured data were collected by the IMP (Isolated Measurement Pods by Solartron Metrology) data acquisition system with frequency of 1.5 Hz.

Figure 2 shows the horizontal test channel (heated length, 508.0 mm; inner diameter, 2.0 mm; wall thickness, 0.5 mm), which is a single circular tube made of nickel GH3128 (China). To measure the local outside wall temperature, the 11 K-type thermocouples (TC) with outside diameter of 0.2 mm were spot-welded on the channel uniformly with 50.0 mm space between each other. The fuel

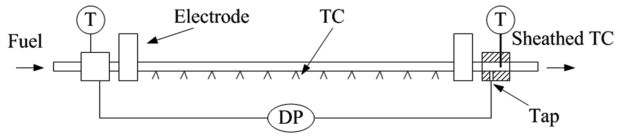


Figure 2. Schematic figure of the electrically heated test section.

temperature was measured in the inlet and outlet by K-type sheathed thermocouples of 1.0 mm outside diameter. The channel was insulated to minimize heat losses and guarantee the accuracy of the wall temperature measurement. Attention must be paid to the distribution of pressure tapping point, which should be put at both ends of the test channel closely approaching the electrodes to avoid any influence of the protruded sheathed thermocouples.

The tube with ID of about 2.0 mm was selected as test channel, for improving the accuracy and repeatability of the test. More uncertainty of pressure drop might be present in a test tube with diameter less than 2.0 mm due to the limited precision of tube manufacture.

Hydraulic resistance due to coke deposition is a time-dependent value. A test run was started by first setting the desired flow rate and outlet pressure. Then, the required heat flux was imposed on the test channel. Finally, the inlet fuel temperature was brought up until the outlet fuel temperature got to the desired condition and then was maintained at the steady state for 30 min.

3. ANALYSIS METHODS

The kerosene kind hydrocarbon fuel (provided by CNPC) is made up of a blend of hydrocarbons, with cycloalkanes occupying 30.50 wt %, alkanes 20.06 wt %, and aromatics 49.42 wt %, with average molecular formula of $C_{11.9}H_{23.4}$, and density of 828.4 kg/m^3 at the condition of 25 °C and atmospheric pressure.

Hydraulic resistance method (HRM) is introduced to find the amount of coke deposition after pyrolysis of hydrocarbon fuel. Thickness of the deposit in test channel was also quantitatively measured by the visualization method (VM). Comparison of the results of the two methods was conducted to verify the feasibility and accuracy of the HRM.[17]

3.1. Hydraulic Resistance Method (HRM). Figure 3 shows a schematic view of the object investigated in HRM. The

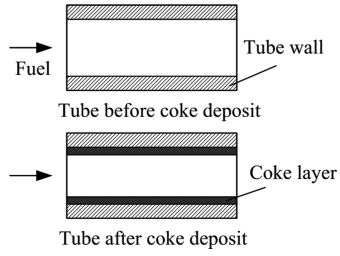


Figure 3. Object investigated in HRM: a single tube before and after coke deposition.

thickness of coke layer is assumed to be uniform along the test channel. As a result of the coke formation, the flow area constricts and the effective diameter decreases. Hydraulic resistance before and after the coke deposition are measured at normal temperature and pressure. The equivalent thickness of the coke deposit (D_e) can be deduced from the increased hydraulic resistance. Though the heterogeneous coke profile is not taken into account, the D_e from HRM is found to be convenient for evaluation of coking property of hydrocarbon fuel and can directly reflect the hydraulic effect of coke deposition in the channel.

dx.doi.org/10.1021/ef3008079 | Energy Fuels 2012, 26, 3672−3679

For the impressible fluid flow in a horizontal channel, the hydraulic resistance loss ΔP is showed in eq 1, where the fluid velocity $V = 4m/(\rho\pi D^2)$. L and D denote the length and inner diameter of the test tube, respectively; m and ρ denote the mass flow rate and density of the fluid. The pressure drops ΔP are measured in the fully developed flow, and then, the Darcy friction factors λ can be determined by eq 2.

$$\Delta P = \lambda \frac{L}{2D}\rho V^2 \tag{1}$$

$$\lambda = \frac{\rho\pi^2 D^5}{8Lm^2}\Delta P \tag{2}$$

For the laminar flow, the friction factor is independent of the relative roughness with ranges of 0 to 0.05,[18] seen in Moody's diagram.[19] For fully developed laminar flow in circular tube, the friction factor from classical theory is used as eq 3. The Reynolds number Re is expressed as $Re = \rho VD/\mu$, where μ denotes the viscosity of the fluid.

$$\lambda = 64/Re \tag{3}$$

Then, the frictional pressure drop can be rearranged as eq 4. At a constant fluid condition (m, ρ, and μ included), C is a constant value, and the pressure drop is inversely proportional to the fourth power of the effective diameter, which provides the fundament principle of HRM. The effect of constricted diameter due to coke deposition is magnified, as a result that a small amount of coke deposition can be reflected in the pressure drop.

$$\Delta P = \frac{128Lm\mu}{\rho\pi D^4} = \frac{C}{D^4} \tag{4}$$

The experimental procedures of HRM are described as follows: (1) Hydraulic resistance before coke deposition was measured for the test channel at normal temperature and pressure. Fuel mass flow rate was controlled at laminar flow condition. (2) Pyrolysis of hydrocarbon fuel was operated at steady state for about 30 min. A specific fuel condition was given for every coking test. (3) Hydraulic resistance was once again measured at the same condition as described in (1). Attention should be paid to guarantee the accuracy of hydraulic resistance measurement, such as eliminating the coke deposits near the pressure taps. For convenience, the hydrocarbon fuel rather than other fluid-like water is used to measure the hydraulic resistance.

3.2. Visualization Method (VM). After the test for HRM, the test channel was cut into short sections (length, 4 mm) at each thermocouple point. The dimensions of test channel and the coke layer were measured by the optical microscope (OM) and scanning electron microscope (SEM) visualization. The optical micrographs were taken and put into the computer by a camera with 8 mega pixels resolution.

Figure 4 shows the optical micrographs of the cross sections of test channel at different TC points with heterogeneous deposit thickness. The blank pipe is also present for contrasting. The coke layer is thin at the inlet but clearly visible at the outlet sections, and the circumferential profiles are nearly uniform on the internal surface. The deposit thickness was measured in the computer, and the processing could be seen from Venkatesan's literature.[17]

The deposit thickness ranges from about several to above 100 μm. The uncertainty of the measured deposit thickness would not exceed 10 μm. These micrographs were also used to measure the inside diameter of test channels with uncertainty of no more than 2.1%.

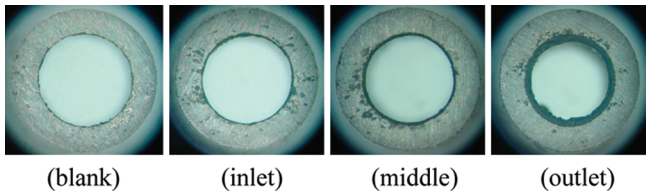


Figure 4. Optical micrographs of the cross section of test sections after the coking test.

4. DATA REDUCTION OF HRM

4.1. Moody Diagram. The average roughness value (Ra) of test channel was measured by a confocal laser scanning microscope. For blank channel without coke deposition, Ra is 0.8 μm, and after coke deposition, the Ra is no more than 20.0 μm. The relative surface roughness is in the range of far below 0.05 (about 100.0 μm) before and after coke deposition. Thus, the Reynolds number is the effect that laminar friction factor only depends on, and the surface roughness has no significant influence.

The friction factors before and after coke deposition are deduced by eq 2. In Figure 5, the friction factors are plotted as a function of Reynolds number (Re), known as Moody diagram. The coking test was conducted at fuel outlet temperature t_b of about 675.0 °C.

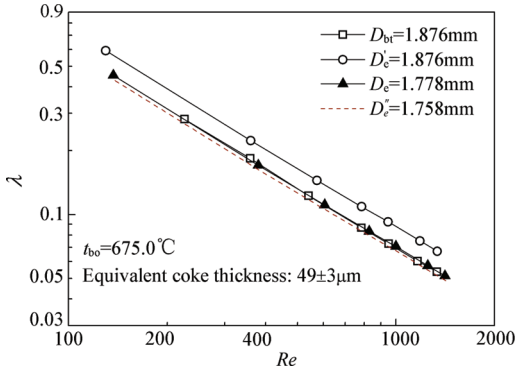


Figure 5. Laminar friction factor vs Reynolds number. D_{bt} is the tube diameter before coke deposition; D_e is the equivalent diameter after coke deposition.

Assuming that the equivalent diameter D_e did not change after coke deposition (the after test diameter D_e equaled to the before test one $D_e' = D_{bt} = 1.876$ mm), experimental results show that the friction factor curve after coke deposition (the hollow circle one) is obviously not the same as the before test one (the hollow square one). However, the friction factor at laminar flow is correlated with Re. It means that there is only one friction factor value at a given Re before and after coke deposition., so the assumption is contradicted. The friction factor was recalculated by reducing the equivalent diameter until the curves of before and after coke deposition were in accordance to each other. The solid triangle curve was attained and agreed well with the before coke deposition one at D_e = 1.778 mm. The equivalent deposit thickness is 49 μm.

The uncertainty of equivalent deposit thickness would not exceed 3 μm in the data reduction by the HRM method. The

dx.doi.org/10.1021/ef3008079 | Energy Fuels 2012, 26, 3672−3679

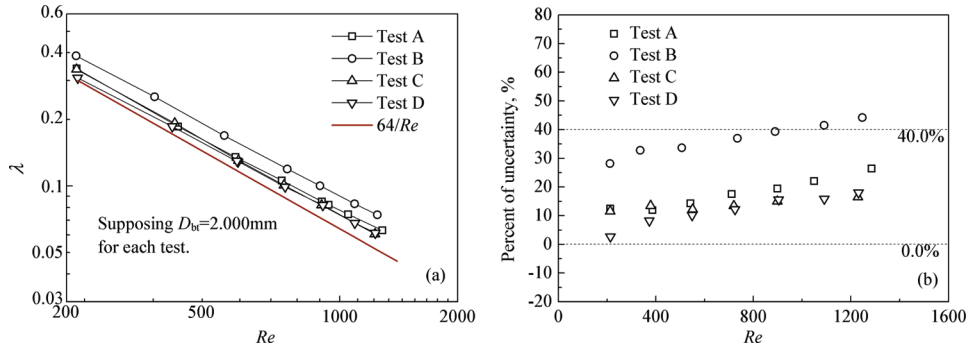


Figure 6. Comparison of the friction factor curves before coke deposition with the theoretical values of 64/*Re*, channel diameters were considered as 2.000 mm.

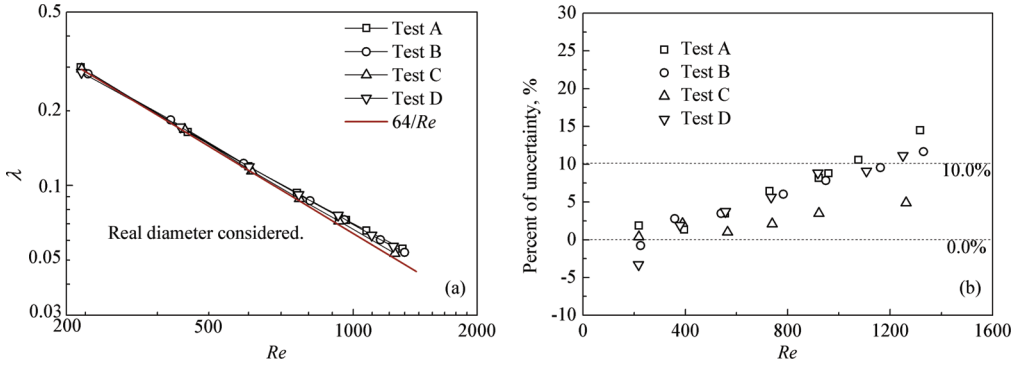


Figure 7. Comparison of the friction factor curves before coke deposition with the theoretical values of 64/*Re*; channel diameters were considered as real measured values.

curve with D_e'' = 1.758 mm is also shown in Figure 5. It can be seen that the curve with uncertainty of 10 μm clearly deviates from the before deposition one. In the Moody diagram, more distance between the before and after deposition curve indicates larger thickness of the deposit.

4.2. Uncertainty Analysis. The position of the friction factor curves on the Moody diagram directly decides the derived equivalent diameter of test channel. To ensure and improve the precision of the HRM, attention should be paid to the following two parameters. As seen in eq 2, the first one is the channel inside diameter before coke deposition, which could affect the calculated friction factor, and the other one is ambient temperature, which affects the fluid properties during the hydraulic resistance measurement.

Considering four test channels with nominal diameter of 2.000 mm, the friction factor curves before coke deposition were compared with the theoretical values of $\lambda = 64/Re$, as shown in Figure 6. Uncertainties of the friction factors exceed 40%. The real channel inner diameters of test A to D are 1.951, 1.876, 1.948, and 1.970 mm, respectively, measured by optical microscope visualization as described above. Test channel B with the smallest inner diameter of 1.876 mm has the largest deviation.

Taking the inside diameter as real measured values, Figure 7 shows each friction factor curve closely approaches the theoretical one. Uncertainties would not exceed 10.0% while *Re* is below 1200, and the uncertainties at the same *Re* are nearly the same with each other.

Fuel temperature is an important parameter to calculate the fiction factors and *Re*. To avoid the effects of temperature in the experimental test, Barlak[20] investigated the friction factor for water flow in microtubes using the circulated bath to maintain the fluid temperature at 25.0 °C. In our paper, the pressure drop measurement was performed at ambient temperatures ranging from about 10.0 to 25.0 °C, where the viscosity of the hydrocarbon fuel greatly decreased from 2.60 to 1.92 mPa·s. The fuel properties are referred to in Hu's thesis.[24]

Figure 8 shows the Moody diagram for different fuel temperatures. It was found that the laminar flow transition to

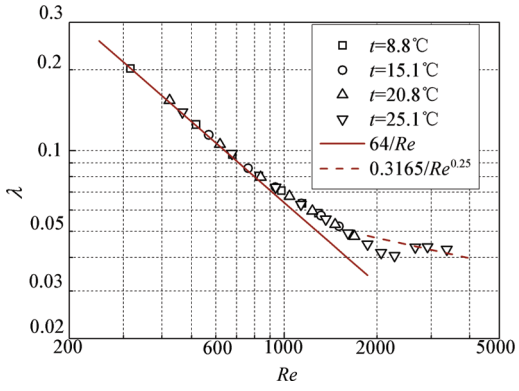


Figure 8. Friction factor as a function of *Re* for different fuel temperatures.

turbulence occurred at *Re* = 2200 at the test channel, using hydrocarbon fuel as working fluid. The transition *Re* is in accordance with the conventional results. The turbulent flow friction factors agree with the Blasius correlation $\lambda = 0.3165/Re^{0.25}$.

The *L*/*D* ratio was selected as above 300 in this paper. For hydrodynamically developing laminar flow in circular tube, the

dx.doi.org/10.1021/ef3008079 | *Energy Fuels* 2012, 26, 3672–3679

relation for the hydrodynamic developing length, L_{hy}, is commonly accepted as $L_{hy}/D = 0.05Re$.[21] When this equation is used for present work, the L_{hy}/D ratio is in the range of 15−110 depending on Re in the whole laminar flow region. Good agreement between the calculated friction factors and the theoretical values verified that the developing laminar flow did not have an important effect on the friction factor, which is consistent with Barlak's[20] results.

The minor loss contribution at the entrance and exit of the test channel can also be ignored in this paper. Thus, the measurement values of pressure drop can be directly used to calculate the fully developed laminar flow friction factors.

In HRM, the Moody diagram based on $200 < Re < 1000$, where the laminar friction factors are best consistent with $64/Re$, was used to evaluate the equivalent thickness of coke deposition. The channel inside diameter before deposition D_{bt} can also be obtained by comparing the measured friction factor with that one in Figure 8. Uncertainties of the HRM are summarized in Table 1. The equivalent deposit thickness H would be no more than (4.1%+3 μm). For a 50 μm coke layer, the uncertainty is 10.1% (5 μm).

Table 1. Uncertainties of the HRM

parameters	uncertainties
diameter, D (%)	±2.1
mass flow rate, m (%)	±1.2
pressure drop, ΔP (kPa)	±0.25
friction factor, λ (%)	±5.1
equivalent diameter, D_e (%)	±2.3
Reynolds number, Re (%)	±2.4
deposit thickness, H	±(4.1% + 3 μm)

5. RESULTS

An experimental investigation on the coking characteristic was conducted at the real fuel system condition of regenerative cooling structure in hypersonic vehicles. The fuel outlet temperature t_{bo} of test channel was in the range of about 650 to 725 °C; heat flux q was about 1250.0 kW/m^2. Mass flux G was about 680.0 kg/m^{2}s, and outlet pressure was about 4.00 MPa. The average real values at steady state for 30 min are shown in Table 2 in detail. At the given heat flux and mass flux, hydrocarbon fuel was preheated before entering the test channel to attain different fuel outlet temperatures.

Table 2. Specific Parameters of Operating Condition for Each Coking Test

test	t_{bo}, °C	t_{bi}, °C	q, kW/m^2	G, kg/(m^{2}s)
A	648.7	16.0	1255.2	693.6
B	676.4	102.5	1247.4	675.5
C	698.0	277.4	1211.8	683.0
D	725.2	339.4	1230.5	680.1

5.1. Results of VM (Visualization Method). For tests A−D, the deposit thickness measured by VM at each TC point is presented in Figure 9a. Results show that coke deposition mostly concentrates at the second half of test channels. Edwards[8] investigated the surface deposit of Jet A-1 and F-T fuel at fuel temperature of about 670 °C by the carbon burn-up method; the profiles of coke deposition are consistent with our results.

The maximum values of coke quantity gradually shift forward with the increasing fuel outlet temperature. The coke deposition is caused by the complicated effect of fuel temperature and channel surface temperature. The inside wall temperatures along the test channel are shown in Figure 9b. Generally, more coke deposition was produced at higher fuel temperature and wall temperature. At the channel entrance, coke deposition was also found, especially at tests A and B. At the case, higher wall temperature occurred at the lower fuel temperature due to the heat transfer deterioration. At tests C and D, the heat transfer deterioration disappeared as a result of the increasing fuel inlet temperature.

The coke deposition was definitely found along the whole channel. Near the outlet of channel coke, deposition decreased at tests C and D. However, no obvious changes of the wall temperature existed. It will be analyzed later, and coke deposition as a function of channel wall temperature and fuel temperature will also be discussed.

5.2. Comparison of Results of HRM and VM. In VM, the average deposit thickness was calculated throughout the whole channel. Results of HRM and VM are compared in Figure 10. The deposit thickness profiles of both methods are identical at different fuel temperatures; the discrepancies are 6.8% on average. Results of HRM are larger than those of VM at high temperatures with more deposit thickness.

The discrepancies of the two methods are mainly contributed to two reasons. First, the simple model of HRM assumes that the deposit thickness profile along the channel is uniform but heterogeneous at the real conditions. Second, the removal of coke may occur during the pressure drop test and the post-test processing, which directly affect the accuracy of the VM. However, the coke deposit, formed at high velocity gas flow, would not probably be removed at the liquid laminar flow.

The consistent results indicate that the HRM was feasible to evaluate the coking characteristic of hydrocarbon fuels. The HRM is an effective method to evaluate onboard the quantity of coke deposit. The pressure drop tests are conducted on the original channel without being destroyed, which is significant, especially for the huge amount of evaluation in selection of an appropriate hydrocarbon fuel with better coking property.

Now in our laboratory, the HRM is being applied well to the evaluation of coking characteristic of hydrocarbon fuels. Compared with the previous literature,[15,16] the HRM developed is a significant milestone to quantitatively correlate the hydraulic resistance with coke deposition.

In addition, the pressure drop measurement is not limited by the structure of the test channel. The HRM can be applied not only on a single channel but also on channels in a flat plate heat exchanger or scramjet.

6. DISCUSSION

6.1. Hydraulic Effect of Coke Deposition during Fuel Pyrolysis. Coke formation accumulates during the pyrolysis process. The pressure drop due to coke deposition is a time-dependent value as a result of the reducing equivalent diameter and increasing surface roughness (for plain tube: 0.8 μm; after coke deposition: 5.0−12.0 μm).

Figure 11 shows the pressure drop, fuel outlet temperature, pressure, and mass flow rate as a function of time during the 30 min coking test at steady state. At the time of 10−40 min, the test run was maintained at steady state. The fuel temperature, pressure, and mass flow rate were controlled at a given value with few discrepancies. Contributing to the coke deposition,

dx.doi.org/10.1021/ef3008079 | Energy Fuels 2012, 26, 3672−3679

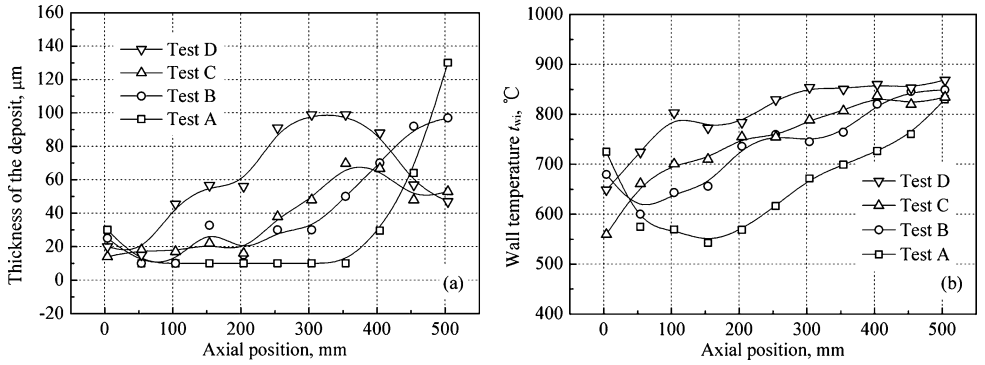


Figure 9. Profiles of (a) deposit thickness and (b) wall temperature along the test channel at different operation conditions.

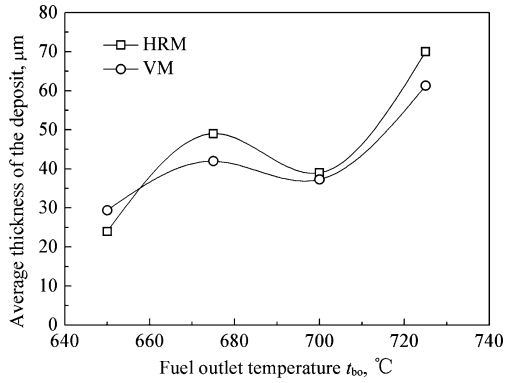


Figure 10. Comparison of results of average deposit thickness throughout the test channel by HRM and VM.

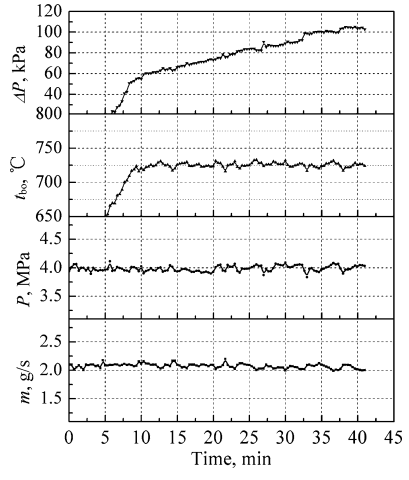


Figure 11. Profiles of pressure drop ΔP, fuel outlet temperature t_{bo}, pressure P, and mass flow rate m as a function of time during fuel pyrolysis at steady states for 30 min at test D. At the time of 10 to 40 min, the test run is stationary; t_{bo} = 725.2 ± 6.9 °C, P = 3.99 ± 0.10 MPa, and m = 2.072 ± 0.077 g/s.

the pressure drop gradually rose from 60 to 105 kPa during the stationary test run. It shows that the hydraulic resistance is significantly affected by coke formation during pyrolysis of hydrocarbon fuel. The more coke deposition, the larger is the pressure drop produced.

6.2. Coking Property and Its Thermal Effect. It was supposed that the coke deposition was a behavior of flowing fluid near the wall. Film temperature,[22] average of the wall temperature, and the bulk fluid temperature are introduced to explain the thermal induced coke formation.

The deposit thickness as a function of film temperature is presented in Figure 12. Results show that the abundant coke

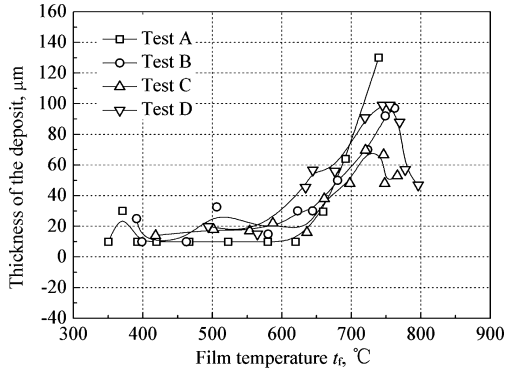


Figure 12. Deposit thickness as a function of film temperature.

deposition commences and increases rapidly at film temperature of about 600 °C. While the film temperature increased to about 750 °C, the deposit approached the maximum value and began to decrease with the increasing film temperature.

Coke layer is always considered as a thermal resistance between the fuel and channel surface, which can cause the heat transfer reduction or even deterioration. Figure 13 shows the behavior of inside wall temperature during fuel pyrolysis at steady states. The increment of wall temperature begins to increase obviously at film temperature of about 650 °C and decrease at about 750 °C. At film temperature above 650 °C,

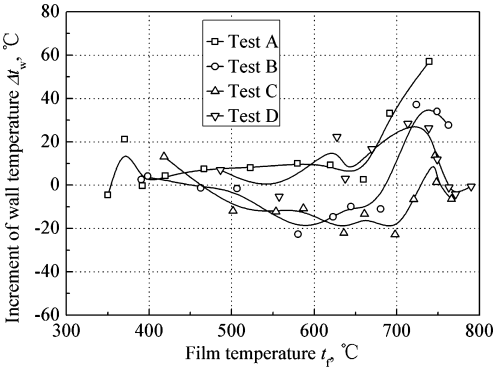
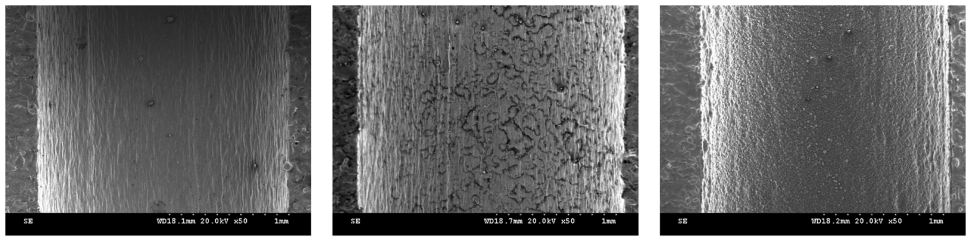


Figure 13. The profiles of increment of wall temperature along the test channels.

dx.doi.org/10.1021/ef3008079 | *Energy Fuels* 2012, 26, 3672−3679



(a) (b) (c)

Figure 14. SEM images of test sections. Magnification is about 50×. (a) t_b = 96.8 °C, t_w = 686.7 °C, and Re = 1976; (b) t_b = 290.2 °C, t_w = 612.9 °C, and Re = 5866; (c) t_b = 563.2 °C, t_w = 726.9 °C, and $Re > 10^4$.

the deposit thickness and increment of wall temperature both increase with the film temperature increasing. Coke deposition as a thermal resistance reduces the heat transfer between hydrocarbon fuel and channel surface.

It is noticed that there are a considerable amount of TC points where the wall temperatures decrease. At these TC points, the coke deposition, instead of reducing heat transfer, increases the local heat transfer coefficient. Figure 14 shows the SEM images of test sections at different positions. It is found that the heat transfer strongly depends on the nature of coke deposition.

At the positions with low deposition rate, the flow turbulence at high Re causes the gully configuration of coke shown in Figure 14b, which enhances the heat transfer. In the enhanced heat transfer region, the gully coke nature was found at fuel temperatures of 165.1 to 479.8 °C (film temperature of 404.6 to 593.5 °C). At the position with high deposition rate of above about 20 μm, the gully coke disappeared, shown in Figure 14c. The nearly uniform coke particles cover the whole channel surface. At the low Re position, the gully coke was not shaped yet, shown in Figure 14a.

The roughness of coke surface was also measured. The Ra (average roughness value) increased up to about 7.5 μm after coke deposition, while the deposit thickness had a range of 10−20 μm. Higher roughness of up to 20 μm can occur at higher deposit thickness (20 μm< H < 130 μm). The friction factor can be hardly affected by the roughness of coke surface, but the convective heat transfer can be profoundly affected.

In summary, the effects of coke deposition on heat transfer depend on two contrary factors: (1) coke deposition increases the roughness of the wetted surface and thereby enhances the heat transfer; (2) coke deposition as a thermal resistance reduces the heat transfer. The two aspects play a crucial role at the low and high deposition rate position, respectively.

To our knowledge, little literature has reported the heat transfer enhancement by coke deposition. The phenomenon was seen in Stiegemeier's presentation[7] and NASA's report[25] without explanations. Previous literature[26] investigated the effect of deposition on the augmentation or reduction of heat transfer. In the recent work by Gascoin,[27] dodecane pyrolysis was investigated experimentally in the stainless steel and titanium reactors; the thermal and hydraulic effect of the coke layer formation was also evaluated, and it suggested that the reduction of flow cross section was an important parameter. Generally, the effect of thermal resistance or roughness depends on the deposition type and channel geometry.

Coking reaction temperature effect is primarily a fluid dynamic effect near the reactor walls.[23] The fuel linear velocity increases as the film temperature is increased. Pyrolysis of endothermal hydrocarbon fuel commences at about 500 °C, and obvious gas products begin to produce at about 600 °C, companied with the increasing coke deposition. The detention time is always a function of coke deposition. The more detention time at high temperature, the more coke deposits, and the detention time is reversed with the fuel velocity. At the film temperature above 750 °C, the deposit thickness together with the increment of wall temperature begins to decrease, as a result of the increasing fuel linear velocity.

7. CONCLUSION

In the paper, the hydraulic resistance method (HRM) was introduced to quantitatively evaluate the coke deposition after pyrolysis of endothermal hydrocarbon fuel in a mini-channel. The HRM mainly considered the hydraulic effect of coke deposition at normal temperature and pressure. It is an onboard and nondestructive method and convenient to carry out on the original test channel. A series of efforts were taken to facilitate the successful implementation of the HRM. The coke layer thickness at each TC point was measured by optical microscope visualization, which was used to validate the feasibility and accuracy of the HRM. Hydraulic and thermal effects of coke deposition during fuel pyrolysis were discussed last.

It was concluded that: (1) The HRM was a feasible and effective way to evaluate the amount of coke deposition in mini-channels. Good agreement was obtained between the results of HRM and optical microscope visualization. The HRM has the unique advantage that coke evaluation is executed onboard without the test channel being destroyed. (2) Coke deposition was profoundly correlated with film temperature during fuel pyrolysis. The great coke deposition commenced and increased rapidly at film temperature of about 600 °C, approached to a maximum value at about 750 °C, and then began to be reduced with the increasing film temperature due to the expansion fuel linear velocity. (3) During the 30 min coking test at steady state, the hydraulic resistance continuously increased in the test channel. Meanwhile, depending on the deposit thickness, the thermal effects of coke deposit can be contrary. At high deposition rate, the coke layer as a thermal resistance reduced the heat transfer. However, at low deposition rate, the gully configuration of coke deposit at high Re turbulence flow can enhance the heat transfer. Now, the HRM has been well applied in the evaluation of coking property of hydrocarbon fuel in mini-channels, and it will be potentially used to evaluate the coking effects in the practical fuel system with the serpent channels in the flat plate heat exchanger or even scramjet in the future.

dx.doi.org/10.1021/ef3008079 | Energy Fuels 2012, 26, 3672−3679

AUTHOR INFORMATION

Corresponding Author

*Tel: +86-029-82665287. Fax: +86-029-82665287. E-mail: qcbi@mai.xjtu.edu.cn.

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

The work is funded by the Open Foundation of the Science and Technology on Scramjet Laboratory of CASIC in China. The financial support of the sponsor is acknowledged.

REFERENCES

(1) Bergholz, R. F.; Hitch, B. D. *AIAA* **1992**, 0515.
(2) Edwards, T. *AIAA* **2003**, 6946.
(3) Mansoori, G. A. *J. Chin. Inst. Chem. Eng.* **2002**, *33*, 25−31.
(4) Zabarnick, S. *Ind. Eng. Chem. Res.* **1994**, *33*, 1348−135.
(5) Heneghan, S. P.; Zabarnick, S.; Ballan, D. R.; Harrison, W. E. *AIAA* **1996**, 0403.
(6) Linne, D. L.; Meyer, M. L.; Edwards, T.; Eitman, D. A. *AIAA* **1997**, 3041.
(7) Stiegemeier, B.; Meyer, M. L.; Taghavi, R. *AIAA* **2002**, 3873.
(8) Edwards, T.; DeWitt, M. J.; Shafer, L.; Brooks, D.; Huang, H.; Bagley, S. P.; Oña, J. O.; Wornat, M. J. *AIAA* **2006**, 7973.
(9) Čižek, Z.; Borek, P.; Fiala, J.; Bogdain, B. *Microchim. Acta* **1990**, *102*, 163−170.
(10) Subudhi, M. M.S. Thesis, The University of British Columbia, Vancouver, Canada, 2006.
(11) Weingarten, J. S.; Euchner, J. A. *SPE Prod. Eng.* **1988**, *3*, 121−126.
(12) Abdumula, F. M. Ph.D. Thesis, University of Miskolc, Miskolc, Hungary, 2005.
(13) Rached, W.; Sicard, F.; Lafargue, A.; Thorel, D. *Int. J. Refrig.* **2007**, *30*, 1393−1400.
(14) Grozdek, M.; Khodabandeh, R.; Lundqvist, P. *Exp. Therm. Fluid Sci.* **2009**, *33*, 357−370.
(15) Gascoin, N.; Gillard, P.; Bernard, S.; Bouchez, M. *Fuel Process. Technol.* **2008**, *89*, 1416−1428.
(16) Chen, A. Y.; Dang, L. *AIAA* **2002**, 0005.
(17) Venkatesanm, M.; Das, S. K.; Balakrishnan, A. R. *Exp. Therm. Fluid Sci.* **2011**, *35*, 531−541.
(18) Kandlikar, S. G.; Schmitt, D.; Carrano, A. L.; Taylor, J. B. *Fluids* **2005**, *17*, 100606.
(19) Moody, L. F. *ASME Trans. J. Appl. Mech.* **1944**, *66*, 671−683.
(20) Barlak, S.; Yapici, S.; Sara, O. N. *Int. J. Therm. Sci.* **2001**, *50*, 361−368.
(21) Steinke, M. E.; Kandlikar, S. G. *Int. J. Therm. Sci.* **2006**, *45*, 1073−1083.
(22) Linne, D. L.; Meyer, M. L.; Braun, D. C.; Keller, D. J. *AIAA* **2000**, 3128.
(23) Edwards, T.; Anderson, S. D. *AIAA* **1993**, 0806.
(24) Hu Z. H. M. S. Thesis, Xi'an Jiaotong University, Xi'an, China, 1996.
(25) Faith, L. E.; Ackerman, G. H.; Henderson, H. T. *NASA CR* **1971**, 72951.
(26) Ceylan, K.; Kelbaliyev, G. *Appl. Therm. Eng.* **2003**, *23*, 557−570.
(27) Gascoin, N.; Abraham, G.; Gillard, P. *J. Thermophys. Heat Transfer* **2012**, *26*, 57−65.