Numerical investigation of pyrolysis effects on heat transfer characteristics and flow resistance of n-decane under supercritical pressure

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1. Introduction

Scramjet engines suffer from extremely harsh thermal circumstances, and thus thermal protection to the scramjet engine is becoming a major consideration for hypersonic aircraft. Regenerative cooling has been considered as the most effective and practical method.1,2 The successful implementation of this
application can lead to greatly improved coolant quality on one hand, and the desired atomization for further efficient combustion on the other hand.³

Hydrocarbon fuel is normally injected into the cooling channel under supercritical pressure. During the regenerative cooling process, the hydrocarbon fuel transfers from subcritical to supercritical state as being continuously heated. As a result, the thermal properties would undergo drastic changes when the fuel temperature approaches its pseudo-critical value. Pyrolysis can make the hydrocarbon fuel absorb an extra amount of heat. Meanwhile, the proportions of the fuel components are changed during pyrolysis. All these factors make the flow and heat transfer phenomenon, particularly the heat transfer characteristics, become extremely complicated.⁴,⁵

Many studies have been devoted to the flow and heat transfer of hydrocarbon fuel under supercritical pressure over the past decades. Hua et al.⁶ numerically studied the effects of inlet pressure, inlet velocity, wall heat flux, and the inlet fluid temperature on the supercritical n-heptane heat transfer processes. It was stated that conventional empirical Gnielinski expression could only be used for supercritical heat transfer predictions of n-heptane under very limited operating conditions. Zhou et al.⁷ experimentally investigated the mechanism of the heat transfer deterioration of RP-3 at supercritical pressure flowing through vertically downward miniature tubes and a reliable RP-3 heat transfer correlation was developed accordingly. Deng et al.⁸,⁹ conducted an experimental study on RP-3 kerosene in a vertical tube at supercritical pressure. It was found that both the heat flux and flow direction influence the heat transfer characteristics due to buoyancy effect. Urbano and Nasuti¹⁰ carried out a parametric numerical analysis on heat transfer deterioration of supercritical methane, ethane and propane. The correlations for estimating the conditions for the onset of heat transfer deterioration were achieved. We¹¹ numerically investigated the characteristics of flow resistance and heat transfer of RP-3 at supercritical pressure and it was stated that pressure drop characteristics become diverse under different pressures with a supercritical temperature. Liu et al.¹² performed an experimental study of the convection heat transfer of n-decane at supercritical pressure in vertical tubes. Their results indicated that the variation of both heat transfer of RP-3 in a micro cooling tube at a supercritical pressure, the fluid mixture are calculated and incorporated in the numerical simulation. The pyrolysis process, convective heat transfer and flow resistance under different conditions, as well as the interaction among them, are analyzed in detail.

2. Numerical methods

2.1. Governing equations and solution method

The cylindrical coordinate form of the governing equations for species continuum, momentum, energy and the k-ε turbulence equation can be written as

Continuity equation:

$$\frac{\partial \rho}{\partial t} + \frac{1}{r} \frac{\partial}{\partial r} \left( r \rho u \right) + \frac{1}{r} \frac{\partial}{\partial \theta} \left( r \rho v \right) + \frac{\partial}{\partial z} \left( \rho w \right) = 0$$

Momentum equation in the axial and radial directions:

$$\frac{1}{r} \frac{\partial}{\partial r} \left( r \rho u \frac{\partial u}{\partial r} \right) + \frac{1}{r^2} \frac{\partial}{\partial \theta} \left( r \rho u \frac{\partial u}{\partial \theta} \right) + \frac{\partial}{\partial z} \left( \rho u \frac{\partial u}{\partial z} \right) = \frac{1}{r^2} \frac{\partial}{\partial r} \left[ \tau_u \left( \frac{\partial u}{\partial r} \right) \right] + \frac{1}{r} \frac{\partial}{\partial \theta} \left[ \tau_u \left( \frac{\partial u}{\partial \theta} \right) \right] + \frac{\partial}{\partial z} \tau_u \left( \frac{\partial u}{\partial z} \right)$$

Energetics equation:

$$\frac{1}{r} \frac{\partial}{\partial r} \left( r \rho C_p \frac{\partial T}{\partial r} \right) + \frac{1}{r} \frac{\partial}{\partial \theta} \left( r \rho C_p \frac{\partial T}{\partial \theta} \right) + \frac{\partial}{\partial z} \left( \rho C_p \frac{\partial T}{\partial z} \right) = \frac{1}{r^2} \frac{\partial}{\partial r} \left( \kappa \frac{\partial T}{\partial r} \right) + \frac{1}{r} \frac{\partial}{\partial \theta} \left( \kappa \frac{\partial T}{\partial \theta} \right) + \frac{\partial}{\partial z} \left( \kappa \frac{\partial T}{\partial z} \right)$$

Turbulence kinetic energy equation:

$$\frac{1}{r} \frac{\partial}{\partial r} \left( r \kappa \frac{\partial k}{\partial r} \right) + \frac{1}{r} \frac{\partial}{\partial \theta} \left( r \kappa \frac{\partial k}{\partial \theta} \right) + \frac{\partial}{\partial z} \left( \kappa \frac{\partial k}{\partial z} \right) = \frac{1}{\kappa} \left[ \frac{\partial}{\partial r} \left( \frac{\tau_u}{\rho} \frac{\partial k}{\partial r} \right) + \frac{1}{r} \frac{\partial}{\partial \theta} \left( \frac{\tau_u}{\rho} \frac{\partial k}{\partial \theta} \right) + \frac{\partial}{\partial z} \left( \frac{\tau_u}{\rho} \frac{\partial k}{\partial z} \right) \right] + \frac{1}{3} \left( \frac{\partial u}{\partial r} \right)^2 + \frac{1}{3} \left( \frac{\partial u}{\partial \theta} \right)^2 + \frac{1}{3} \left( \frac{\partial u}{\partial z} \right)^2$$

Energy flux and heat transfer characteristic processes of the fluid mixture are calculated and incorporated in the numerical simulation. The pyrolysis process, convective heat transfer and flow resistance under different conditions, as well as the interaction among them, are analyzed in detail.
where $p$ is the pressure, $\mu$ is the viscosity, and $g$ is the acceleration of gravity.

Energy equation:

$$\frac{\partial (\rho u H)}{\partial x} + \frac{\partial (\rho v H)}{\partial y} = \frac{\partial}{\partial x} \left( \lambda \frac{\partial T}{\partial x} \right) + \frac{\partial}{\partial y} \left( \lambda \frac{\partial T}{\partial y} \right) - \frac{\rho v H}{r} + \frac{\lambda}{r} \frac{\partial T}{\partial r} + S$$

where $H$ is the specific enthalpy, $T$ is the temperature, $k$ is the thermal conductivity, and $S$ is the energy source term due to chemical reaction.

$k$-$\varepsilon$ turbulence equation:

$$\frac{\partial (\rho u k)}{\partial x} + \frac{\partial (\rho v \varepsilon)}{\partial y} = \frac{\partial}{\partial x} \left( \mu + \mu_t \frac{\partial k}{\partial x} \right) + \frac{\partial}{\partial y} \left( \mu + \mu_t \frac{\partial \varepsilon}{\partial y} \right) - \frac{\rho v \varepsilon}{r} + \frac{1}{r} \left( \mu + \mu_t \right) \frac{\partial \varepsilon}{\partial r} + G_k - \rho \varepsilon$$

$$\frac{\partial (\rho u \varepsilon)}{\partial x} + \frac{\partial (\rho v \varepsilon)}{\partial y} = \frac{\partial}{\partial x} \left( \mu + \mu_t \frac{\partial \varepsilon}{\partial x} \right) + \frac{\partial}{\partial y} \left( \mu + \mu_t \frac{\partial \varepsilon}{\partial y} \right) - \frac{\rho v \varepsilon}{r} + \frac{1}{r} \left( \mu + \mu_t \right) \frac{\partial \varepsilon}{\partial r} + C_1 G_k \frac{\varepsilon}{k} + C_2 \rho \varepsilon^2$$

where $k$ is the turbulent kinetic energy, $\varepsilon$ is the dissipation rate of $k$, $\mu_t$ is the eddy viscosity, and $G_k$ is the turbulent production due to buoyancy and shear stress. $C_1$, $C_2$, $\sigma_k$ and $\sigma_\varepsilon$ are constants.

The governing equations were solved by the Computational Fluid Dynamics (CFD) software FLUENT. The SIMPLEC algorithm was utilized for the pressure field, and the second-order upwind scheme was used to discretize convection terms. The $k$-$\varepsilon$ two-equation model was adopted to simulate the turbulent flow and the “enhanced wall treatment” was employed to determine the near wall velocity, which could well predict the convective heat transfer in the tube.18,21,22

2.2. Chemical kinetics model

A one-step PPD chemical model was proposed by Ward et al.16 based on their experimental measurements. The measured mass fractions of the major decomposed products at different pressures were averaged to obtain a general reaction mechanism.16

$$\text{C}_{10}H_{22} \rightarrow 0.151\text{H}_2 + 0.143\text{CH}_4 + 0.256\text{C}_2\text{H}_4 + 0.126\text{C}_2\text{H}_6$$
$$+ 0.230\text{C}_3\text{H}_6 + 0.180\text{C}_3\text{H}_8 + 0.196\text{C}_4\text{H}_8$$
$$+ 0.102\text{C}_4\text{H}_{10} + 0.171\text{C}_5\text{H}_{12} + 0.124\text{C}_6\text{H}_{14}$$
$$+ 0.195\text{C}_6\text{H}_{12} + 0.089\text{C}_7\text{H}_{14} + 0.169\text{C}_7\text{H}_{16}$$
$$+ 0.072\text{C}_8\text{H}_{16} + 0.152\text{C}_8\text{H}_{18} + 0.012\text{C}_9\text{H}_{18}$$
$$+ 0.053\text{C}_9\text{H}_{18} + 0.003\text{C}_9\text{H}_{20}$$

(7)

The pyrolytic reaction rate of n-decane can be calculated as

$$\frac{d(C_{10}H_{22})}{dt} = -k_c(C_{10}H_{22})$$

(8)

![Fig. 1](Fig. 1) Validation of calculated thermophysical properties of n-decane.
where the rate constant \( k_c \) is computed using the Arrhenius expression

\[
k_c = A_c e^{-E/(RT)}
\]  

(9)

The pre-exponential factor \( A_c = 1.6 \times 10^{13}/s \) and the activation energy \( E = 263.7 \text{ kJ/mol} \), the universal gas constant \( R = 8.314 \text{ J/(mol·K)} \).

2.3. Thermophysical property calculations

The calculation of the density \( \rho \), viscosity \( \mu \) and thermal conductivity \( \lambda \) for n-decane and each pyrolytic product are based on the extended corresponding states principle. Propane is selected as the reference material, whose state equation adopts 32 parameters Modified Benedict-Webb-Rubin (MBWR) equation.\(^{23}\) For each species and the mixture, the density is achieved according to the corresponding relation between the species and the reference propane, and the viscosity and the thermal conductivity are derived from the TRANsport Property Prediction (TRAPP) method,\(^{23}\) and the thermal capability \( c_p \) is calculated by means of deviation function method.

Fig. 1 shows the density, specific thermal capacity, thermal conductivity and viscosity of n-decane under different supercritical pressures calculated by the methods above and the data of National Institute of Standard and Technology (NIST). Good agreement has been reached between the calculated results and the data of NIST.

2.4. Computational model and mesh

Fig. 2 shows the schematic of the physical model of the flow in a circular tube with the length of \( L = 900 \text{ mm} \) and the diameter of \( d = 2 \text{ mm} \). From Fig. 2, it demonstrates a 600 mm heated section enforced with a constant and uniform surface heat flux. At the inlet, a adiabatic section with 150 mm in length is included to ensure a fully developed flow, while at the outlet a adiabatic section with 150 mm in length is adopted to avoid the outflow boundary effects. The computational model is modeled in a 2D axial-symmetric domain. The pyrolysis behavior of n-decane is influenced by the pressure and wall heat flux. The operating pressure ranged from 3 to 5 MPa, with different wall heat fluxes from 1.0 to 1.2 MW/m\(^2\). The inlet temperature was 400 K, and mass flow rate remained constant at 2 g/s. All these geometric parameters and the boundary conditions are relevant to the practical regenerative cooling applications in the scramjet. Meanwhile, special heat transfer characteristics, such as heat transfer deterioration and enhancement, could be well observed under these parameters.

A mesh independence study was conducted to identify an appropriate mesh density for the aimed calculations. In the current work, a set of computational mesh of 900 \( \times \) 40 in the axial and radial directions has been proved to be sufficient, and the radial mesh is exponentially fined in the near-wall region. Under the current computational conditions, \( y^+ \) of the first near-wall node is kept at approximately 1.0, which is required by the “enhanced wall treatment”.

3. Results and discussion

3.1. Validation of chemical kinetics model and numerical method

In the current study, the openly published experimental and computational data of Ward et al.\(^{16}\) were used to validate the developed chemical kinetics model and numerical method. In accordance with the experimental conditions, the tube length and diameter are 0.375 and 0.5 mm respectively. The inlet temperature \( T_{in} \) is 473 K, and the outlet pressure \( p \) is 3.45 MPa. The mass flow rate ranges from 0.25 to 0.75 mL/min, with different wall heat fluxes of 1.0, 1.1 and 1.2 MW/m\(^2\). In the current work, the n-decane conversion is defined as

\[
y = (1 - Y_{C_{10}H_{22}}) \times 100\%
\]  

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The location at $x/d = 0$ starts from the beginning of the heated section in the following discussion. As illustrated in Fig. 4(a), the n-decane conversion under 5 MPa maintains close to zero before $x/d = 150$, since pyrolysis does not normally occur. After $x/d = 150$, the n-decane conversion starts to increase and increasing rate rises drastically. Fig. 4(b) reveals that n-decane conversion begins to increase at $x/d = 45$ under 3 MPa. The increasing rate of n-decane conversion tends to slow down from $x/d = 100$, which is followed by a second rise after $x/d = 200$, as shown in Fig. 4(b). The n-decane conversion increase at the beginning of the heated section under 3 MPa becomes more obvious when the wall heat flux goes up.

Fig. 5 shows the detailed distribution of temperature and logarithm reaction kinetic rate in the symmetry plane of the heated section under the heat flux of 1.2 MW/m². The variation of the temperature near the wall indicates that heat transfer deterioration appears before $x/d = 100$, and it could be due to the drastic variation of the physical properties. As can be seen in Fig. 5(b), the wall temperature reaches about 1100 K before $x/d = 100$. As the reaction rate is highly sensitive to the temperature, it is the extremely high pyrolysis rate near the wall, which gives rise to n-decane conversion increase at the beginning of the heated section, as shown in Fig. 5(d). As illustrated in Fig. 5, heat transfer deterioration disappears and turns to heat transfer enhancement in the segment of $100 < x/d < 200$. The decreased wall temperature reduces the pyrolysis rate near the wall. In addition, pyrolysis would not normally occur with the main stream temperature that is below 750 K. Therefore, the n-decane conversion increases slowly in heat transfer enhanced segment. After $x/d = 200$, the main stream temperature and pyrolysis rate continuously go up, and accordingly, bring about a sharp rise of the n-decane conversion.

Heat transfer deterioration would be less evident or even disappear with the increasing pressure, as the physical property changes become smaller under a higher pressure. Fig. 6 demonstrates the distribution of bulk fluid temperature $T_f$ and wall temperature $T_w$ under 5 and 3 MPa. With a much smaller degree of heat transfer deterioration, the wall temperature under 5 MPa is much lower than that under 3 MPa before $x/d = 100$. This explains the late increase of n-decane conversion under 5 MPa, in contrast with that under 3 MPa.

It can be also seen from Figs. 4(a) and (b) that the n-decane conversion at $x/d = 100$ is about 0.5% under 5 MPa and is about 4% under 3 MPa. After $x/d = 100$, the conversion under 5 MPa increases by 37% and reaches about 37.5% at the end of the heated section, while that under 3 MPa increases by 34.5% and eventually reaches about 38.5%. The fluid temperature reaches 600 K at $x/d = 100$, as illustrated in Fig. 6. The density decreases slower under 5 MPa than that under 3 MPa when the
temperature is above 600 K, as shown in Fig. 1(a). As the smaller density gives rise to the larger acceleration, the residence time of the fluid flowing from \(x/d = 100\) to the end of the heated section is shorter under 3 MPa. This accounts for the larger increase of conversion after \(x/d = 100\) under 5 MPa.

### 3.3. Pyrolysis effects on heat transfer characteristics

Fig. 7 shows the heat transfer performance along the heated section of the tube under the condition of \(p = 3\) MPa, \(q = 1.2\) MW/m², and two sets of result, with and without consideration of pyrolysis, are compared. The n-decane conversion results in this case can be referred to from Fig. 4(b). Fig. 7(a) shows the distribution of the bulk fluid temperature and the wall temperature, and Fig. 7(b) shows the distribution of the heat transfer coefficient which is defined as

\[
h = \frac{q}{T_w - T_l}
\]

As illustrated in Fig. 7, at the beginning of the heated section, the increase of the heat transfer coefficient with pyrolysis occurs much earlier than that without pyrolysis. Accordingly, pyrolysis shortens the heat transfer deteriorated segment and lowers the maximum value of the wall temperature. After \(x/d = 150\), the results with pyrolysis deviate from the results without pyrolysis, due to the increasing amount of reacted n-decane. The higher heat transfer coefficient and the lower fluid bulk temperature with pyrolysis make the wall temperature much lower than that without pyrolysis. As shown in Fig. 7(a), the fluid temperature and wall temperature are reduced by 90 and 110 K at the end of the heated section respectively.

Figs. 8 and 9 show the detailed distribution of the physical properties and velocity in the symmetry plane of the heated section with and without pyrolysis respectively, under the same condition of Fig. 7. As the tube flow is heated up, the temperature near the wall quickly exceeds the pseudo-critical temperature. Because of the descending density and specific thermal capacity, the heat capacity near the wall decreases, which would lower the heat transfer capability and lead to extremely high temperature near the wall.\textsuperscript{10,25,26} After the flow is further heated up, the increasing bulk temperature leads to the growth of velocity. The increased flow velocity causes the recovery of the coolant capability, and consequently enhances the heat transfer process.\textsuperscript{10,25} As revealed by the comparison between Figs. 8 and 9, pyrolysis brings about little change in thermal conductivity, and makes the fluid heat capacity lower due to the increasing proportions of the small molecule productions. However, the heat transfer process with pyrolysis is less deteriorated before \(x/d = 150\) and much more enhanced after \(x/d = 150\), as shown in Fig. 7. This indicates that the further fluid acceleration caused by pyrolysis plays the dominant role in convective heat transfer. Figs. 8(c) and 9(c) demonstrate the larger velocity with pyrolysis. At the end of the heated section, the flow velocity without pyrolysis is about 13 m/s, while that with pyrolysis reaches 20 m/s. It is the larger velocity with pyrolysis that leads to the higher heat transfer coefficient.

As revealed from the results under heat fluxes of \(q = 1.0, 1.1\) MW/m² in Fig. 10 and \(q = 1.2\) MW/m² in Fig. 7, pyrolysis shortens the heat transfer deteriorated segment and lowers the temperature maximum to a larger extent, as the heat flux becomes greater. The temperature maximum without pyrolysis rises from 1051 to 1167 K, while that with pyrolysis rises from 1030 to 1126 K when the heat flux increases from 1.0 to 1.2 MW/m². After \(x/d = 150\), the wall temperature tends to be constant at the end of the heated section as the heat flux becomes greater. It can be explained by the fact that the greater heat flux could lead to larger conversion which restrains the fluid temperature from going up and enhances the convective heat transfer. The temperature reached at the
end of the heated section without pyrolysis rises from 1118 to 1230 K, whereas that with pyrolysis rises from 1081 to 1109 K when the heat flux increases from 1.0 to 1.2 MW/m². From the above analysis, it can be concluded that pyrolysis is conducive for the cooling structure to withstand a higher heat flux under a certain temperature allowed by solid material.

3.4. Pyrolysis effects on flow resistance

It is well known that friction on the tube wall causes pressure drop along the axial direction. The axial pressure gradient can be used to indicate the flow resistance, seen in Eq. (14):

$$\frac{dp}{dx} = f \frac{1}{d} \frac{\rho u^2}{2} = f \frac{G}{2A} u$$  \hspace{1cm} (14)

where $f$ is the resistance coefficient.

Fig. 7 Heat transfer characteristics with/without pyrolysis ($p = 3$ MPa, $q = 1.2$ MW/m²).

Fig. 8 Distribution of physical properties and velocity with pyrolysis ($p = 3$ MPa, $q = 1.2$ MW/m²).

Fig. 9 Distribution of physical properties and velocity without pyrolysis ($p = 3$ MPa, $q = 1.2$ MW/m²).

Fig. 11(a) shows the distribution of axial pressure gradient under the condition of $p = 5$ MPa, $q = 1.2$ MW/m² with and without pyrolysis. The pressure gradient without pyrolysis has little variation after $x/d = 150$, while that with pyrolysis begins to increase abruptly. As discussed in detail in the previous study, the dramatic variation of supercritical fluid density plays the dominant role in pressure drop. As can be clearly seen from Fig. 11(b), the further reduced density caused by pyrolysis leads to larger acceleration. The greater radial velocity gradient near the wall enlarges the shear stress and friction at the wall. That explains why pressure gradient increases abruptly and becomes different from that without pyrolysis.

Fig. 12(a) shows the distribution of axial pressure gradient under the condition of $p = 3$ MPa, $q = 1.2$ MW/m² with and without pyrolysis. In contrast with the case under 5 MPa, the differences caused by pyrolysis begin to appear at $x/d = 45$, etc.
and pressure gradient with pyrolysis is little greater in the section from \( x/d = 45 \) to 105. This results from the earlier occurrence of pyrolysis, which leads to a more rapid acceleration in this section, as shown in Fig. 12(b). Moreover, the value of pressure gradient under 3 MPa is much greater than that under 5 MPa, attributing to the lower density and higher velocity under 3 MPa.

4. Conclusions

In this paper, a numerical investigation of n-decane flow and heat transfer coupled with pyrolysis is conducted. The heat transfer and the pyrolysis process under supercritical pressure, as well as the mechanism of pyrolysis effects on heat transfer characteristics and flow resistance, are discussed and analyzed.
Numerical investigation of pyrolysis effects on heat transfer characteristics and flow resistance

in detail. Based on the results of the current research, the following specific conclusions may be made:

(1) The pyrolytic reaction rate is highly sensitive to the temperature, and therefore, the n-decane conversion arises rapidly as the fluid temperature becomes high enough after \( x/d = 150 \). The n-decane conversion before \( x/d = 150 \) remains nearly to be 0 under 5 MPa, but has an increment under 3 MPa. This contrast is caused by the heat transfer deterioration under 3 MPa.

(2) Pyrolysis gives rise to a further decrease in fluid density, which leads to a further acceleration. The enhanced heat transfer and the lower fluid temperature jointly slow the rise of wall temperature, and the value of the wall temperature is much lower than that without pyrolysis.

(3) Fluid acceleration plays the dominant role in heat transfer enhancement. Pyrolysis in the heat transfer deteriorated segment makes the fluid velocity higher, which results in an earlier rise of the heat transfer coefficient. As a consequence of this, pyrolysis shortens the heat transfer deteriorated segment and lowers the temperature maximum.

(4) The further acceleration of fluid caused by pyrolysis plays an important role in bringing about much greater flow resistance. Attributing to the higher velocity, pressure gradient increases abruptly as pyrolysis takes place after \( x/d = 150 \).

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