

Technical Analysis of Thermal Decomposition Characteristics of Liquid Hydrocarbon Fuels for a Regenerative Cooling System of Hypersonic Vehicles

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Abstract

A technological review and analysis were performed on thermal cracking of aviation hydrocarbon fuels that circulate as coolants in regenerative cooling systems of hypersonic flights. Liquid hydrocarbons decompose into low-carbon-number hydrocarbons when they absorb a considerable amount of energy at extremely high temperatures, and these thermal cracking behaviors are represented by heat sink capacity, conversion ratio, reaction products, and coking propensity. These parameters are closely interrelated, and thus, they must be considered for optimum performance in terms of the overall heat absorption in the regenerative cooling system and supersonic combustion in the scramjet engine.

Key Words : Scramjet Engine, Endothermic Reaction, Aviation Hydrocarbon, Pyrolysis, Catalytic Cracking

1. Introduction

Hypersonic flight vehicles fly at Mach 5 or higher in the Earth's atmosphere [1]. The scramjet engine, which is an air-breathing propulsion engine, is highly efficient and effective because it has a high specific impulse characteristic compared with the rocket propulsion engine and operational flexibility in a wide range of speeds from supersonic to hypersonic [1,2]. However, flying at supersonic speeds using this scramjet engine causes issues such as excessive heat due to aerodynamics on the fuselage and within the engine combustion chamber and effectively mixing and burning the hydrocarbon liquid fuels in complex and supersonic air flow conditions that are accompanied by a shock train. Active regenerative cooling has been studied extensively to address these issues [1,3,4].

An active regenerative cooling system circulates the vehicle fuel where it is needed as a coolant using the endothermic reaction of liquid hydrocarbon fuels [3,5], solving the heating problem of the fuselage and engine caused by hypersonic flight [1]. When this method is applied, the liquid fuels at room temperature and are heated to high temperatures and endothermally decomposed into low-carbon-number hydrocarbons during circulation. Consequently, the combustion efficiency of the hydrocarbon fuels supplied to the supersonic combustion chamber of the scramjet engine can

also be greatly improved [6]. However, when hydrocarbons that have reached a supercritical state are injected into a supersonic combustor of the scramjet engine through the injectors of various shapes and sizes, they exhibit patterns that are completely different from the injection characteristics of ordinary liquid fuels [7]. As a result, these patterns greatly influence the mixing, igniting, and burning of the fuel in the supersonic combustor [8]. Consequently, the operating characteristics of the regenerative cooling system, including the hydrocarbon fuels, become an important variable in determining the overall propulsion performance of the scramjet engine.

The key elements that determine the overall operational performance of a regenerative cooling system of hypersonic flight vehicles include the endothermic decomposition characteristics of the hydrocarbon fuels, flow and heat transfer properties of the cooling channel, and the material and structure of the cooling system. The characteristics of liquid aviation hydrocarbon fuels, which have been used as fuels for air intake propulsion engines for several decades, vary by type. However, as presented in Fig. 1, these fuels can be thermally cracked and pyrolyzed into low-carbon-number hydrocarbons [11,12], while absorbing a large amount of heat at high temperatures [9,10]. Furthermore, when an appropriate catalyst is applied, the hydrocarbons undergo an endothermic reaction more actively than during thermal decomposition and catalytic cracking may occur [11,13].

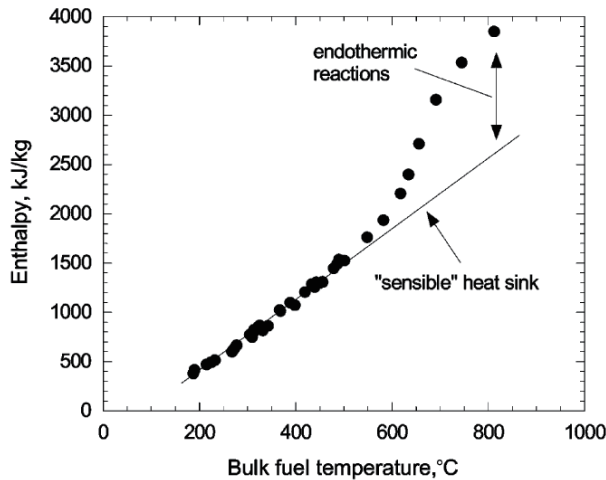


Fig. 1 Heat Sink Capacity from Endothermic Cracking of JP-7 [9,10]

Accordingly, this study, aiming toward the development of an efficient regenerative cooling system for hypersonic flight vehicles, identified global research trends related to the pyrolysis of liquid aviation hydrocarbon fuels and conducted detailed technical analysis.

2. Pyrolysis characteristics of hydrocarbon liquid fuels

The parameters that indicate thermal decomposition characteristics of hydrocarbon aviation fuels that are circulated as a coolant in the regenerative cooling system for hypersonic flight vehicles are divided into heat sink capacity, conversion ratio, product distribution, and coking propensity. The research trends and results of detailed technical analyses for each of these parameters are as follows.

2.1 Heat Sink

Heat sink is the most significant performance parameter of the regenerative cooling system and only occurs as a sensible, or physical, heat sink until the decomposition reaction occurs, as shown in Fig. 1. However, when the reaction temperature reaches a certain temperature and the decomposition reaction begins, the system not only contains a physical heat sink but also a chemical heat sink, which indicates the chemical reaction energy required for pyrolysis. This effect depends on the reaction temperature and pressure of the fuel, residence time, flow rate, and the presence or absence of a catalyst and steam [9,10,14]. JP-7, which has the best thermal stability among the aviation fuels developed so far, kerosene-based JP-8/Jet A-1, which is mainly used as fuel for aircraft, JP-10, which is a high density special fuel, and RP-3, which is kerosene-based aviation fuel used in China, all have a significant level of endothermic performance [9,15]. Table 1

compares the heat sinks of various fuels, tested under similar conditions [9].

Table 1 Comparison of Total Heat Sink Capacity of Several Hydrocarbon Fuels [9]

Fuel	JP-7	JP-8+100	JP-10	n-octane
Total heat sink [kJ/kg]	3415	2908	2605	2493

Figure 2 shows the changes in the chemical heat sink characteristics according to the reaction temperatures and pressures of n-decane, conducted by Zhou et al. [14]. Thermal decomposition begins when the temperature is over 750 K, and the chemical heat sink increases rapidly as the reaction temperature and pressure increase. However, when the temperature rises above 900 K, this trend slows down, and from 920 K, the heat sink decreases according to the pressure. This result occurs because the decomposition reaction of hydrocarbons is converted to an exothermic reaction when it reaches a certain temperature [14]. In particular, the heat sink change and the overall heat sink size are closely related to the conversion and product distribution, as discussed in the following section.

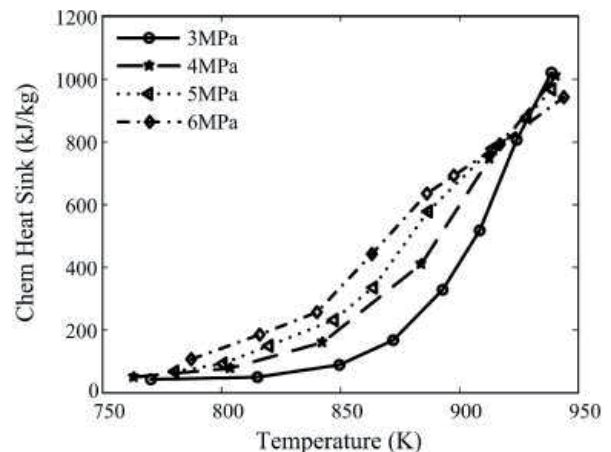


Fig. 2 Chemical Heat Sink Capacity of N-decane Pyrolysis at Various Pressures [14]

2.2 Conversion

Conversion refers to the rate at which the original hydrocarbons are converted into another component. In general, conversion is known to be proportional to temperature, similar to heat sinks [14,16], and accordingly, heat sinks and conversion are also proportional to each other. Fig. 3 illustrates the conversion rate of n-decane according to temperature and pressure, and the conversion increases as the temperature and pressure increase. This trend is similar to that of a heat sink that increases with the temperature and pressure, as shown in Fig. 2.

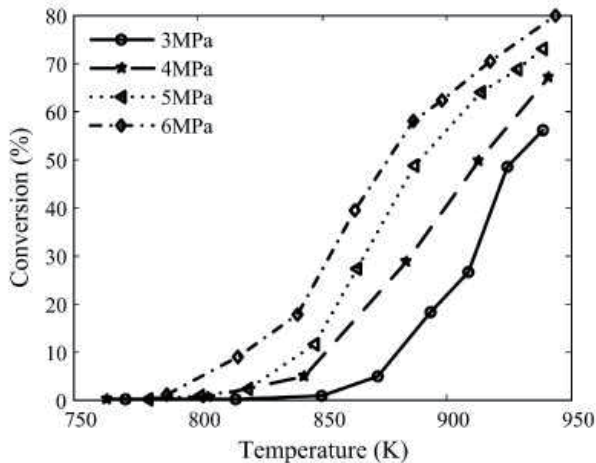


Fig. 3 Percentage of Conversion of N-decane Pyrolysis [14]

However, a high conversion rate is not directly linked with a sizable heat sink. This result can be confirmed in Fig. 4, demonstrating the relationship between the conversion rate of n-decane and the heat sink according to the pressure. According to the figure, when the conversion rate is steady, the heat sink at a low temperature appears larger, which is closely related to the product distribution. That is, when the original hydrocarbon fuel component is decomposed, the conversion unconditionally increases; however, the proportion of the endothermic reaction among the various decomposition reactions varies depending on the reaction conditions even when the conversion remains the same, and the product distribution varies accordingly. This implies that when conversion ultimately increases, the heat sink also generally increases. However, the absolute value of the heat sink is more closely related to the proportion of the endothermic reaction during the decomposition reaction rather than the conversion itself and the resulting product distribution.

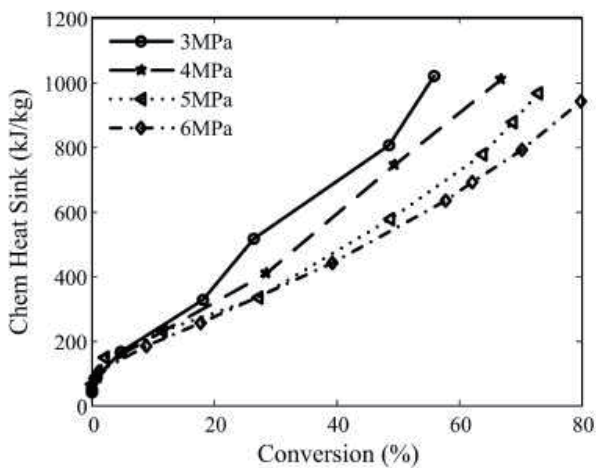


Fig. 4 Chemical Heat Sink at Varying Conversion Rates of N-decane Pyrolysis [14]

Thus, using the results of Fig. 3 and 4 in connection with conversion could help in explaining the phenomenon in which the heat sink changes according to the pressure at temperatures above 920 K is reversed, as observed in Fig. 2. As shown in Fig. 3, the conversion ratio continues to increase as the temperature and pressure increase. However, in Fig. 4, the heat sink at a constant conversion rate is larger when the pressure is lower, and accordingly, in Fig. 2, the conversion at a low pressure and temperature below 920 K is relatively low with a low heat sink. However, when the temperature rises above 920 K, the conversion rate at low pressures increases rapidly, resulting in a reversal in which the heat sink becomes larger.

2.3 Product Distribution

Product distribution, along with the conversion rate, is known to have a significant influence on the heat sink of hydrocarbon fuels as well as the characteristics of the endothermically decomposed fuels supplied to the combustion chamber of scramjet engine, where they are mixed, ignited, and combusted with the supersonic air flow. Accordingly, this parameter is directly connected to the propulsion performance of the scramjet engine. First, from the heat sink perspective, if kerosene ($C_{12}H_{24}$) is completely decomposed into ethylene (C_2H_4), it has a theoretical heat sink of 3.56 MJ/kg. However, it is decomposed into various elements such as CH_4 , C_2H_4 , and C_2H_6 with the corresponding heat sink of approximately 1.2 MJ/kg, which varies considerably from the theoretical value [10]. This result is due to a reactor residence time that is insufficient to cause a complete decomposition reaction to produce significant amounts of paraffinic hydrocarbons. In particular, paraffinic products involve an exothermic reaction, with more produced when the reaction reaches equilibrium. Accordingly, when the reaction time is extended, complete endothermic decomposition that increases the heat sink does not occur. However, the phenomenon of increasing paraffinic hydrocarbon products dominates, and in most cases, the heat sink exhibits a decreasing trend. Moreover, the propensity for the paraffinic product to increase and the heat sink to decrease becomes more pronounced as the reaction pressure increases [10,14,17]. As a result, as seen in Fig. 4 regarding n-decane, as the pressure increases, the heat sink at the same conversion rate decreases. Additionally, in Fig. 2, as the fuel temperature increases, the exothermic reaction in which the saturated hydrocarbon components increase under high pressure conditions dominates, and the heat sink decreases [14,17–19].

The paraffin/olefin ratio, or alkane/alkene ratio, is used as a parameter indicating the trends of the increase in the saturated paraffinic hydrocarbon product and the decrease in heat sink [10, 14]. Based on the results of several studies, as shown in Fig. 5, as the reaction pressure increases, the alkane portion increases and the alkene portion decreases [19]. As a result, the heat sink was found to be directly inversely proportional to the alkane/alkene ratio [10,14,17–19]. Edwards [10]

determined that a short reaction time under high temperature conditions lowers the paraffin/olefin ratio, which is advantageous in increasing the heat sink.

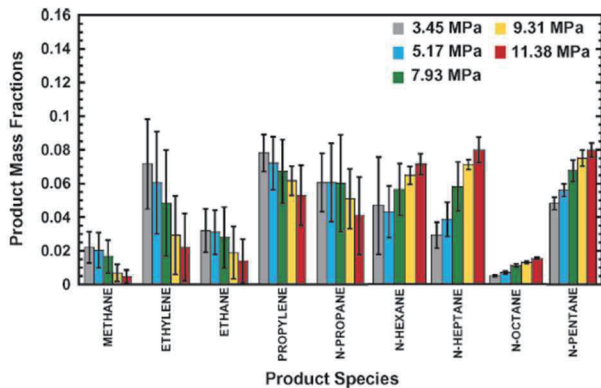


Fig. 5 Product Distributions of N-decane Pyrolysis for Various Pressures [19]

However, the product distribution is directly related not only to the heat absorption characteristics but also to the supersonic combustion performance of the scramjet engine. There is an advantage in improving combustion efficiency when there are more low-carbon hydrocarbon components, such as C_2H_4 , including H_2 , which has a relatively fast flame propagation speed [6]. Fig. 6 presents the comparison results of the endothermically decomposed products of typical hydrocarbon fuels with low molecular weight hydrocarbons with 5 or fewer carbon atoms, including hydrogen, at a specific reaction pressure. Although there are differences for each fuel, H_2 , CH_4 , C_2H_4 , C_2H_6 , and C_3H_6 are diversely distributed. Particularly, these components vary significantly depending on the reaction conditions, such as temperature, pressure, and residence time, and the presence or absence of a catalyst and steam [9,10,14,15,19].

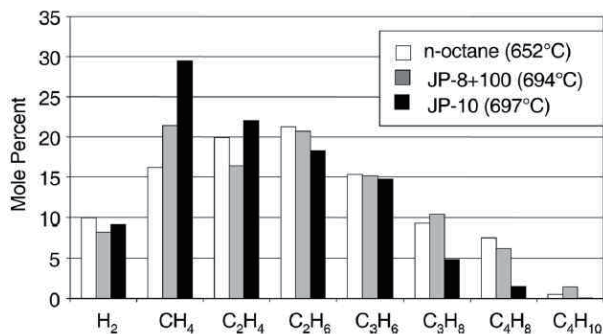


Fig. 6 Comparison of High Pressure Cracking Products (<C5) of Various Hydrocarbon Fuels [9,10]

Moreover, in the case of mild cracking of less than 20% conversion, various studies found that there is a proportional product distribution (PPD) characteristic in which the

components of the product distribution are maintained at a constant ratio regardless of the temperature and conversion, as shown in Fig. 7 [10,19]. Using this result, an endothermic decomposition reaction model of hydrocarbon liquid fuels was developed, and a simulation was conducted and compared with the experimental results. In this respect, the researchers confirmed that a prediction with engineering accuracy for the endothermic decomposition properties is possible, as presented in Fig. 8 [10,19–21].

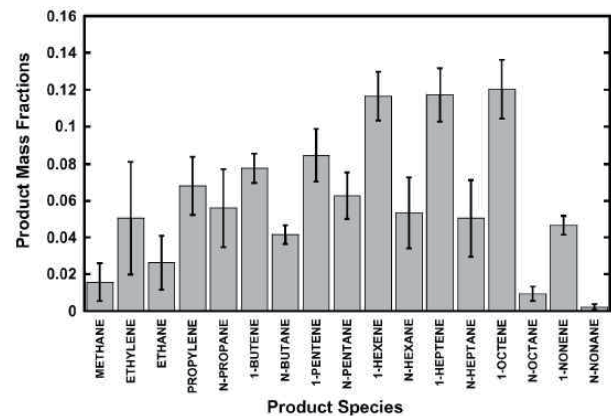


Fig. 7 Averaged PPD of N-decane under Various Pressures [19]

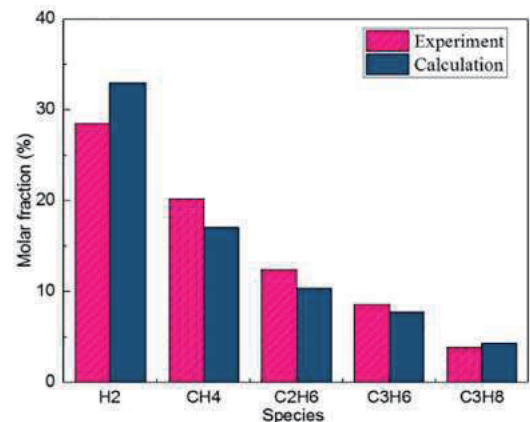


Fig. 8 Comparison of Product Distribution between Experiment and PPD Calculation [20]

2.4 Coking

As shown in the test results for Jet-A fuel in Fig. 9, coking, as a by-product from fuel decomposition, is known to increase exponentially in accordance with the reaction temperature, residence time, and conversion [10,22]. The most significant problem of coking is that the materials are deposited on the surface of the flow path of the regenerative cooling system and inside the fuel injector of the scramjet engine, preventing the flow and heat transfer, potentially blocking the flow path. Accordingly, when discussing the performance of endothermic fuels, not only the overall heat sink but also the ability to

inhibit coking are critical factors [23].

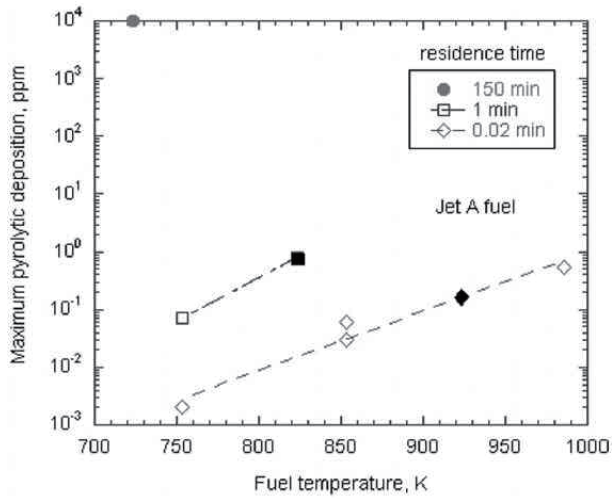


Fig. 9 Peak Pyrolytic Deposition with respect to Fuel Outlet Temperature [10,22]

Coking is divided into filamentous coking and amorphous coking depending on the form generated. Coking was originally identified as a by-product when producing ethylene using naphtha as a raw material in a petrochemical plant [10,24]. Particularly, there are three major mechanisms for coking. Filamentous coking occurs when the hydrocarbons are catalyzed by the metal surface of a reactor. Amorphous coking occurs as liquid high molecular weight carbon components such as tar are deposited through condensation polymerization as the hydrocarbons decompose. In addition, amorphous coking occurs due to vapor-phase poly-condensation when gaseous radicals and low molecular hydrocarbons are deposited on a solid surface [10,25].

This coking phenomenon was confirmed to similarly occur in the endothermic decomposition process of hydrocarbon liquid aviation fuels that circulate in the regenerative cooling system at high temperatures and pressures [10,23]. Sun et al. [23] experimentally compared and analyzed the characteristics of pyrolysis and coking in supercritical conditions according to the composition of the hydrocarbon fuels. In general, filamentous coking has been identified as being detrimental. However, in their study, as shown in the picture on the right in Fig. 10, when filamentous coking was observed on the inner surface of the reactor, it implied that amorphous coking is relatively insignificant, and thus, the ability to inhibit coking is stronger. In other words, when the filamentous coking on the metal surface is completely covered by amorphous (or spherical) coking due to the coking formation process presented in Fig. 11 and becomes invisible (refer to the picture on the left in Fig. 10), it implies that a significant amount of coking occurred [23].

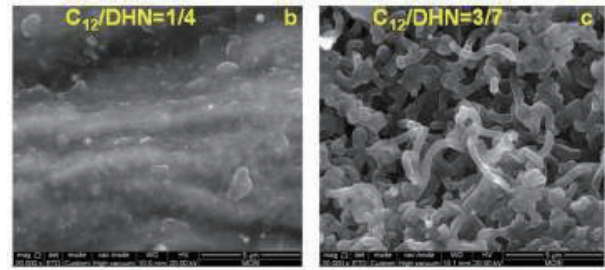


Fig. 10 SEM of Reactor Inner Surface; Flat and Dense Carbon Layer (Left), and Typical Filamentous Carbon (Right) [23]

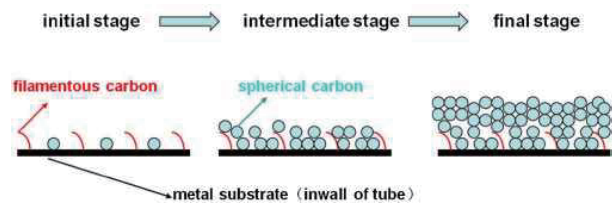


Fig. 11 Process of Carbon Deposition during Pyrolysis [23]

Al or Mn alloys, which resist coking, have been used as the interior material of the reactor of regenerative cooling systems, along with the addition of appropriate channel surface coatings, such as aluminum and silica, to suppress the occurrence of filamentous coking. [10,26]. When steam was involved in the reaction during endothermic decomposition reactions at high temperatures, the overall amount of coking, including amorphous coking, was significantly reduced [10,27–29]. Moreover, when the flow path surface was processed more precisely, less amorphous coking in the form of particles was deposited. Furthermore, Edwards [10] presented the lowest possible reaction pressure, a short residence time in the reactor, and the removal of local hot spots to minimize coking during the endothermic decomposition reaction of hydrocarbon fuels to maximize ethylene production.

However, several studies confirmed that polycyclic aromatic hydrocarbons (PAH) are an intermediate product closely related to the occurrence of coking under high pressure conditions, such as in a regenerative cooling system. These PAHs are rapidly generated as the reaction pressure and fuel conversion increase. In particular, fuel at supercritical temperatures has increased the solubility of high molecular weight components, and thus, even when PAHs are generated, it does not lead to coking. However, when cooled below the critical temperature, PAHs were immediately deposited as carbon [10,30,31].

3. Improvement of endothermic decomposition characteristics and improvement plan

Catalytic cracking and steam reformation technologies have been utilized to improve the endothermic decomposition characteristics of hydrocarbon jet fuels, which are currently being actively studied worldwide. Various studies are being conducted regarding catalytic decomposition to find a method of maximizing the heat sink capacity and conversion at a relatively low temperature and minimizing contamination, which decreases the activity of the catalyst over time. Research regarding the steam-reformation technologies has continued because adding water vapor during the catalytic decomposition of hydrocarbon fuels not only improves the catalytic decomposition properties but also effectively suppresses coking. Accordingly, a detailed analysis on the improvement of the endothermic decomposition characteristics will be performed through subsequent studies.

4. Conclusions

This study identified global research trends related to the pyrolysis of liquid hydrocarbon aviation fuels and conducted a detailed technical analysis for the development of an efficient regenerative cooling system for hypersonic vehicles. Hydrocarbon liquid fuels, which are the most basic element among those that determine the operating performance of the regenerative cooling system for hypersonic flight vehicles, absorb a substantial amount of heat at high temperatures and decompose into low molecular weight hydrocarbons. The main parameters that represent the pyrolysis performance are heat sink, conversion ratio, product distribution, and coking propensity.

Heat sink is directly related to fuel conversion and product distribution. In general, as the conversion increases, heat sink also increases. However, the absolute value of heat sink was found to be more closely related to the proportion of the endothermic reaction during decomposition and the resulting product distribution rather than the conversion itself. Especially when the reaction pressure increases, conversion increases. However, as the temperature increases, exothermic reactions, in which the saturated hydrocarbon component increases, dominates and the heat sink decreases. Thus, although the conversion ratio is not always proportional to the heat sink capacity, high conversion has a great influence on the increase in the combustion performance of the scramjet engine. Accordingly, deriving the reaction conditions in which the heat sink capacity, conversion ratio, and product distribution are optimal is critical in terms of the cooling and propulsion performance of the entire propulsion system in connection with the combustion performance of the scramjet engine. Furthermore, not only the overall heat sink capacity but also the ability to inhibit coking is vital when discussing

the performance of endothermic fuels.

In conclusion, continuous and in-depth studies of various liquid hydrocarbon fuels under various temperature and pressure conditions are required for the development of practical hypersonic vehicles capable of operating in a wide range of speeds.

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