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A THEORETICAL INVESTIGATION ON THERMAL INSULATION OF MULTILAYER PASSIVE THERMAL PROTECTION SYSTEM WITH CARBON PHENOLIC COMPOSITES IN COMBUSTION CHAMBER

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ABSTRACT

In order to understand the thermal insulation performance of a multilayer passive thermal protection system (MPTPS) in combustion chamber, a one-dimensional multilayer transient heat conduction approach has been developed to simulate the thermal insulation procedure in MPTPS. The MPTPS includes carbon phenolic composites which pyrolyze subject to high temperature, leading to the change of its thermal properties, such as density, specific heat capacity and thermal conductivity. Incidentally, the pyrolytic gas generated from composite material infiltrates into the other layers and gaps between them in MPTPS, leading us to take the effects of contact thermal resistance into account. On the other hand, the latent heat results from the phase change of the carbon phenolic composite may cause an extra heat source, which releases or absorbs heat with the variation of time. Consequently, the wall temperature on the outermost layer in MPTPS has been observed and plotted against time. The effects of these factors introduced above have been carefully analyzed in terms of the variation of wall temperature with time.

INTRODUCTION

In view of the development of hypersonic vehicles, ramjet and scramjet engines are expected to be widely applied as the component of providing power. Due to the high requirement of the speed flight and maneuverability, the inner walls of the combustion chamber and nozzle suffer strong aerodynamic heating, which increases especially during the acceleration to cruise phase (Shripad et al., 2015). In order to avoid the engine from damaging, an effective thermal protection of inner walls of combustion chamber and nozzle from the hot gas is definitely needed. The thermal protection system (TPS) may be classified as active thermal protection system, passive thermal protection system and their combination. Since the active thermal protection, such as film cooling, consumes the coolant sucked directly from atmosphere, leading to the deficient of air for combustion. Therefore, in some occasions, the application of passive thermal protection is preferred to achieve higher specific impulse and thrust coefficient (Li et al., 2013).

Passive thermal protection system (PTPS) refers to as a multilayer shield protecting the outer metal wall from the hot gas directly contacting with, maintaining the temperature over the metal wall under a reasonable level. The subject of multilayer thermal protection has been attracted a great interest in many aspects, such as thermal protection layer covers entire surface of the spacecraft and re-entry vehicles (Kumar et al., 2016), multilayer thermal insulations for high-temperature fuel cell (Markus et al., 2013) and passive thermal protection in ramjet combustion. In general, the multilayer passive thermal protection system (MPTPS) is composed of several materials with particular function arranged in a sequence, forming a multilayer coat to resist thermal load from aerodynamic heating or hot gas. The design of MPTPS should concern the properties of material and requirements of thermal insulation. According to the protection, the TPS materials can be categorized as ablative material and insulative material (Xie et al., 2013). Ablative material such as ceramic composites exhibits an excellent performance of withstand high temperature and ablation (Wu et al., 2020), whereas the insulation materials such as carbon-phenolic and thermal protection blankets consist of fibre fibrics and insulation (Davis et al., 1999) behave a lower density and lower thermal conductivity (Ma et al., 2015). The combination of both materials leads to an integrated PTPS, which is able to resist the ablation and to insulate heat flux to the outer metal wall.

An integrated passive multilayer thermal protection usually includes the following components, namely, one layer of ceramic fiber matrix, which is directly suffering the erosion and ablation of hot gas; one layer of carbon-phenolic pyrolytic material, which exerts its advantages of good mechanical and insulative characteristics (Massoud, 2021); one layer of flexible insulation material which has low density, high porosity, lower or graded thermal conductivity. In recent decades of years, many researchers have conducted plenty of studies on ablation characteristic of ceramic fiber matrix, pyrolysis characteristics of carbon-phenolic, insulation design of low thermal conductivity blankets. However, the investigations on heat insulation performance of the whole passive multilayer thermal protection, which concerns the pyrolysis and flow of pyrolysis gas, extra heat generation, are not sufficient. Therefore, in this study, the pyrolysis characteristics of carbon/phenolic materials were experimentally studied, furthermore, the influence of carbon/phenolic materials on the thermal insulation performance of MPTPS subject to high temperature was studied numerically.

EXPERIMENT ON PYROLYSIS CHARACTERISTICS OF CARBON/PHENOLIC MATERIALS

Carbon/phenolic material is a composite material made of carbon fiber and phenolic resin, which will be pyrolyzed under the high temperature and generate pyrolysis gas at the same time. In order to understand the pyrolysis characteristics of carbon/phenolic composite, experiments of tube furnace heating and PY-GC-MS were carried out to determine the mass loss law of carbon / phenolic and the composition of pyrolysis gas. In this work, three groups of carbon / phenolic materials were fabricated, the sizes were characterized by diameter and height, namely, $\varphi \times h$ = 13 * 2mm, 13 * 4mm, 13 * 6mm, respectively, as shown in Figure 1.

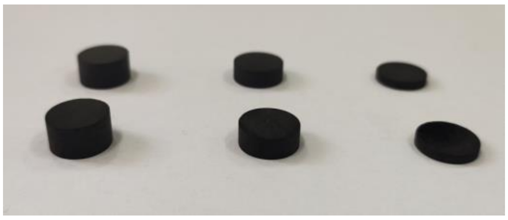


Figure 1 Carbon/phenolic composites

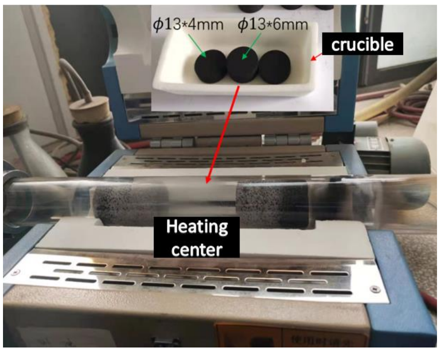


Figure 2 tube furnace heating experiment

PY-GC-MS experiment

Pyrolysis gas generated at different temperatures (300℃, 400℃, 500℃, 600℃, 700℃, 800℃) was captured by pyrolysis gas-mass spectrometer, and gas composition was analyzed by mass spectrometer.

Tube furnace heating experiment

The mass loss of carbon/phenolic material has been investigated by heating the carbon/phenolic samples in a tubular furnace, the mass difference was obtained by measuring its weights before and after heated. As shown in Figure 2, two experimental samples size of φ13 * 4 mm and φ13 * 6 mm (hereinafter referred to as size 1 and size 2) were placed in the crucible and put together in the heating center of the tubular furnace.In the temperature range of 100℃-900℃, different experimental pieces were heated in sections with a heating rate of 10 ℃/min and a residence time of 4 mins. The mass change of the cooled specimen was measured by a balance. Moreover, the surface morphology after pyrolysis was scanned by a high power scanning electron microscope.

NUMERICAL MODEL OF MULTI-LAYER THERMAL PROTECTION STRUCTURE

As introduced above, MPTPS is generally composed of ablation resistant layer, insulation layer and metal shell layer (Kumar et al.,2016; Ding et al.,2021). The carbon / phenolic material selected as one of the layers because of its bearing capacity and lower thermal conductivity. In this study, the finite element method has been adopted to construct the one dimensional numerical model of MPTPS, as shown in Figure 3. The one dimensional transient conduction equation has been integrated and discreted, forming an equation set composed of several matrixs as (1), which can be solved by the gaussian method.

$$[K]\{T\}+[C]\{\dot{T}\}=\{P\} \qquad (1)$$

where the $[K]$ is matrix of thermal conductivity, $[C]$ is matrix of heat capacity, $\{\dot{T}\}$ is matrix of first order time derivative, $\{P\}$ is matrix of thermal load, which may include thermal loads of heat source, the second typy and the third type of thermal boundary conditions.

The material of superalloy GH99 was selected as the metal shell layer, which is needed to be protected from the hot gas. Hence, the ablative material of C-C/SiC was arranged innermost to resist the high temperature and to suffer erosion. The carbon/phenolic composite was arranged just close to the C-C/SiC composite, enhancing the bearing ability of C-C/SiC and resisting heat transfer simultaneously. In addition, a flexible material of aluminium silicate ceramic fiber paper with high porosity and much lower thermal conductivity was placed between caobon/phenolic and outer metal wall. According to the experimental data and some references (Hu et al.,2017; Ma et al.,2015), the parameters of model and properties of each material were given in Table 1.

Table 1 Parameters of MPTPS material

Material layer	Ablation resistant layer	Carbon/phenolic layer	Insulation layer	Metal shell layer
Thickness (mm)	6	12	3	4
Thermal conductivity (W/m-K)	85-45 (22-1800℃)	0.686-1.4 (22-1800℃)	0.04-0.155 (22-1200℃)	8.5-27.21 (22-1000℃)
density （kg/m^3）	1900	1418-1073 (22-1800℃)	200	8470
specific heat capacity （J/kg-K）	763-1700 (22-1800℃)	1000	800	440-700 (22-1000℃)

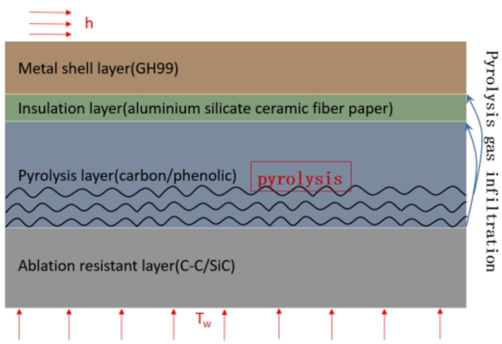


Figure 3 MPTPS numerical model

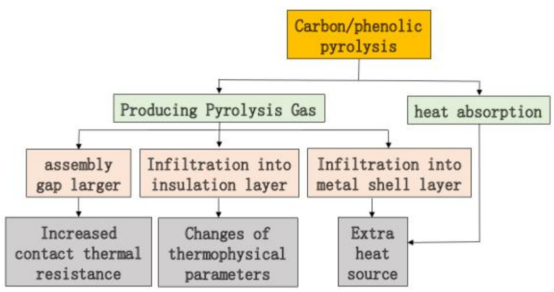


Figure 4 Analysis of factors affecting thermal insulation performance by pyrolysis of carbon/phenolic materials

The pyrolysis of carbon/phenolic composites brings plenty of effects on the thermal insulation performance of MPTPS, as described in Figure 4, the discussion is summerized as follows:

1) Contact resistance between each layer: the effects contact resistance may be more prominent. The deformation of each layer and immersion of pyrolysis gas may result in the arise of gap between two layers, which may leads to an extra thermal resistance. According to (Liu et al.,2010), the value of contact thermal resistance is usually, the corresponding thermal conductivity is 400-1000 w/(m^2•k). However, because the insulation layer is made of porous flexible material, only the most influential interface of carbon/phenolic—C-C/SiC is considered.

2) The change of thermal properties of the insulation layer: the flexible insulation material is a porous structure with high porosity and low thermal conductivity. The infiltration of gas into the flexible insulation material leads to the change of its thermal properties. According to the porous media theory, its equivalent thermal conductivity can be simply calculated by empirical formula (1).

$$k_{eff} = \varepsilon k_f + (1-\varepsilon)k_s \quad (1)$$

Where, k_f and k_s are the thermal conductivities of the air and the solid, respectively. While ε is porosity of the material, i.e., the volume fraction of the pore in material. The immersion of the pyrolysis gas into the material caused the change of k_f, so does the effective thermal conductivity.

3) Internal extra heat source: The pyrolysis of carbon/phenolic material under high temperature produced pyrolysis gas, which is complicated in composition and difficult to predict the amount of heat. Moreover, the pyrolysis gas will penetrate into the layer gaps and pores in insulation materials, leading to the change of thermal contact resistance and thermal properties of insulation material. In the process of thermal insulating, the carbon/phenolic layer directly contact with the ablation layer, which is able to withstand high temperature but with a higher thermal conductivity, leading to the quick increase of temperature of the carbon/phenolic material, in other words, the temperature of carbon phenolic material rises rapidly up to almost the same as that of hot gas. However, the pyrolysis of carbon phenolic will spend some time. On the other hand, in referencing as (SHI et al., 2013), the magnitude of heat release of High silica / phenolic composites under 2200K is about 10^{6}W/m^2. Hence, in present study, in line with ground experiment results, the volumetric heat release rate of carbon phenolic material with the thickness of 12mm has been approximately set as 10^{7}W/m^3 at the time of 600s, while the magnitude of heat flux in equation(3) represents the heat flux of pyrolysis gas flows from carbon phenolic to the outer metal wall was set a smaller value, i.e. 30000W/m^2.

Therefore, in this paper, a random mathematical describtion of heat source has been constructed as shown in formulas (2) and (3) respectively, in which the (2) is the heat absorbtion of pyrolysis, while the formula (3) represents the heat flux of pyrolysis gas flows to the outer metal wall.

$$\Phi(\mathrm{W/m^3}) = -1.67 \times 10^4 t\,, \quad 0 \le t \le 600\mathrm{s} \quad (2)$$

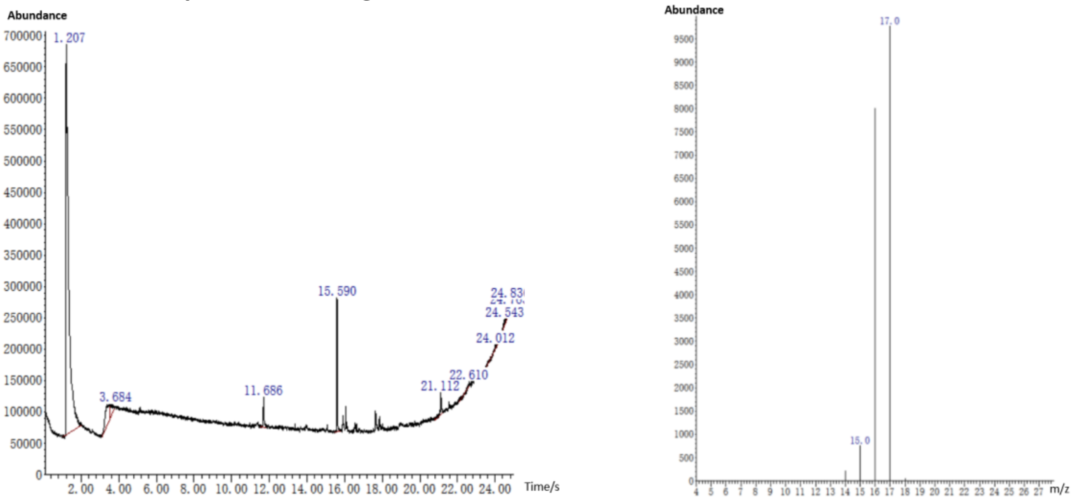
$$\mathrm{q}(\mathrm{W/m^2}) = \begin{cases} 0 & ,0 \le t < 5\mathrm{s} \\ 2000t - 10000 & ,5\mathrm{s} \le t < 20\mathrm{s} \\ 30000 & ,20\mathrm{s} \le t < 30\mathrm{s} \\ -1500t + 75000 & ,30\mathrm{s} < t < 50\mathrm{s} \\ 0 & ,50\mathrm{s} \le t \le 600\mathrm{s} \end{cases} \quad (3)$$

As can be seen in the table 1, the thickness of MPTPS is definitely small, therefore, the total grid number is adopted a small value, i.e., 251. One of the difficulties is dealing with the iteration of thermal property parameters of material, which are the function of temperature. The volumetric heat source (eq.(2)) was imbedded within the layer of carbon phenolic, which can be traced by its coordinate, while the heat flux (eq.(3)) can be imposed on the lower surface of outer metal shell.

RESULTS AND ANALYSIS

PY-GC-MS experimental results

The results of pyrolysis PGC and mass diagram at 300℃ are shown in Figure 5 . According to Figure 5(a), the captured particles with high abundance can be obtained, and then the relative atomic weight can be found from Figure 5(b). Then, the composition can be predicted by matching the mass spectrogram with the material database. The proportion of this component is determined according to the peak area, so as to determine the main components of pyrolysis gas under various temperature conditions. As a result, the components of gas at different pyrolysis temperature were analyzed and determined finally as shown in Figure 6.



(a)PGC diagram **(b) mass spectrum diagram**

Figure 5 PGC and mass diagram of thermal cracking at 300℃

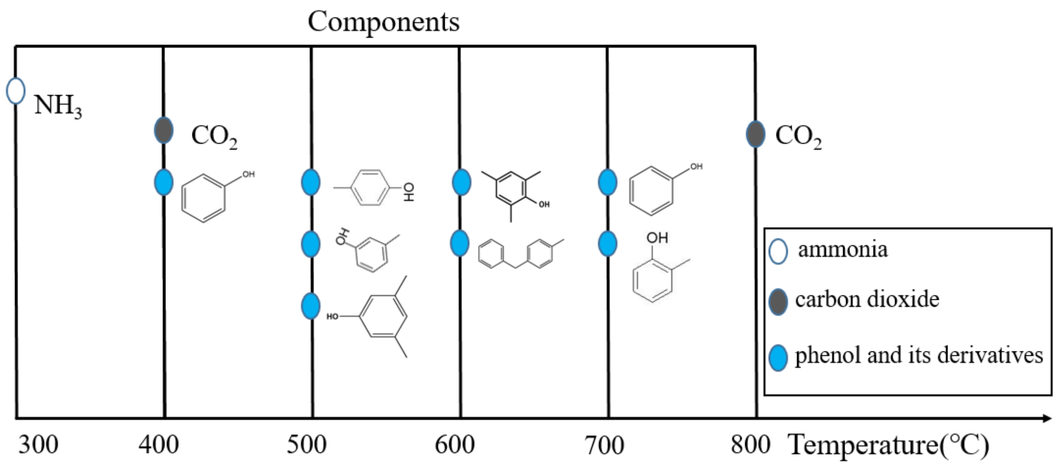


Figure 6 main components of gas at different temperatures in PY-GC-MS experiment

It can be seen from the Figure 6 that the pyrolysis products of carbon/phenolic composites are mainly carbon dioxide and phenol and its derivatives at first. With the increase of temperature, the proportion of carbon dioxide becomes prominent. This is mainly because the ignition temperature of phenol is 715℃, which illustrates that the phenol may be completely burned and converted into carbon dioxide at 800℃.

Tube furnace heating experiment

The experimental results of tube furnace heating are shown in Table 2.

Table 2 Mass change of specimens at different temperatures

Temperature (℃)	Mass (g) (size 1)	Mass (g) (size 2)	Temperature (℃)	Mass (g) (size 1)	Mass (g) (size 2)
Room temperature -126	0.75	1.14	500-600	0.61	0.96
126-208	0.745	1.13	600-700	0.57	0.87
208-302	0.73	1.11	700-800	0.57	0.84
302-402	0.71	1.07	800-900	0.57	0.85
402-500	0.69	1.00	500-900	0.56	-

Taking size 1 as an example, further analysis shows that its mass loss and temperature change are shown in Figure 7.Carbon/phenolic materials begin to pyrolyze at 200℃, and the mass loss is about 2.7% at 400℃ and 500℃, while the maximum mass loss is 11% at 600℃. A large amount of thick smoke comes out from the side nozzle of the tubular furnace with strong coke smell. When the temperature is 700℃, the mass is almost unchanged. This may because after 700℃, the phenolic resin in the composites is almost completely pyrolyzed, and the remaining substances are no longer changed. The final residual mass of the specimen heated continuously (500-900℃) is 0.56g, which is the same as that of the specimen heated by sections, and can be used as verification. Therefore, it can be seen from the experimental results that the carbon/phenolic composite material is heated at high temperature, and only some of its components are pyrolyzed, resulting in mass loss. However, according to the observation of the heated experimental piece, the size of the carbon/phenolic composite material has hardly changed, and only the surface morphology becomes rough due to pyrolysis and oxidation reaction. For this reason, the surface morphology was scanned by high power scanning electron microscope, as shown in Figure8 and Figure 9 .

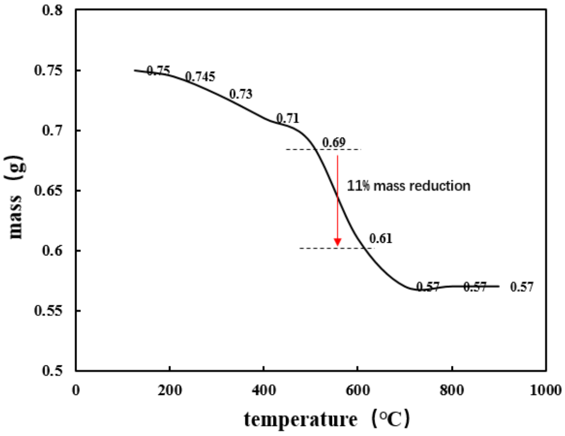
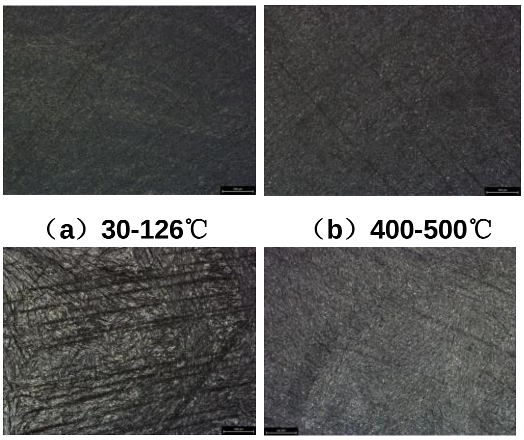
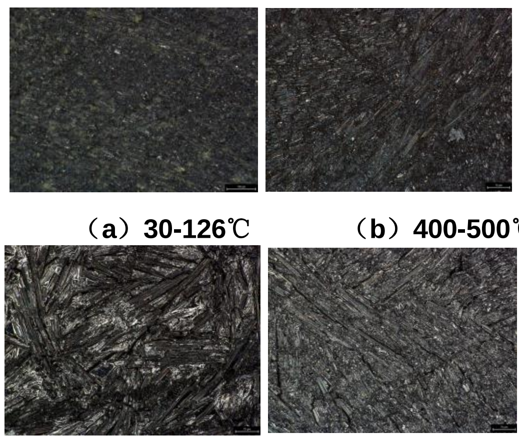


Figure 7 mass loss diagram of carbon/phenolic pyrolysis



（a）30-126℃ **（b）400-500℃**

（c）800-900℃ **（d）500-900℃**



（a）30-126℃ **（b）400-500℃**

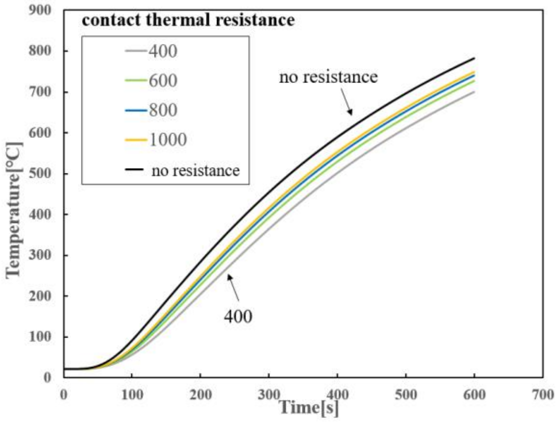
（c）800-900℃ **（d）500-900℃**

Figure 8 100 times pixel scanning **Figure 9 500 times pixel scanning figure**

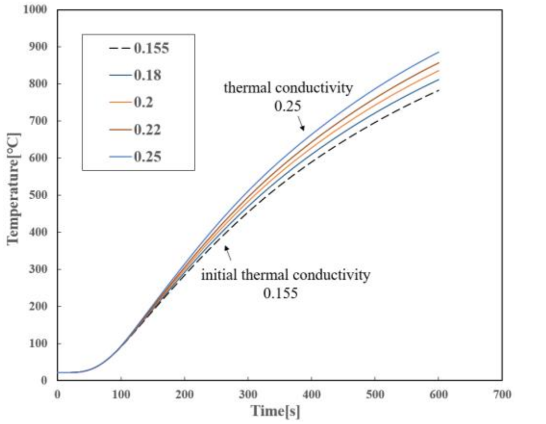
It can be seen from Figure 8 and Figure 9 that even the heating temperature rises to 900℃, the surface of carbon / phenolic only had a pyrolysis of phenolic matrix and some carbon fibers were exposed, while had little effect on the appearance of carbon / phenolic materials.

Influence of pyrolysis of carbon/phenolic materials on thermal insulation performance of MPTPS

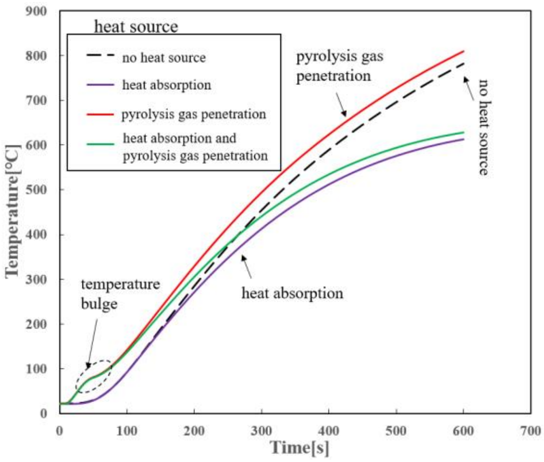
As emphasized above, the influence factors result from the pyrolysis of carbon phenolic material, includes thermal resistance of interlayer gap, the variation of thermal properties of thermal insulation layer and the extra heat source were taken into account.Reference with the environment of ground test, the convective heat transfer coefficient of metal wall with ambient is set to be 20w/(m^2•K), while the inner wall surface suffers a constant wall temperature of 1800°C. Hence, the one-dimensional transient heat conduction problem with extra heat source and thermal contact resistance has been conducted. Finally, the metal wall temperature for different cases were plotted against operation time in Figure 10.



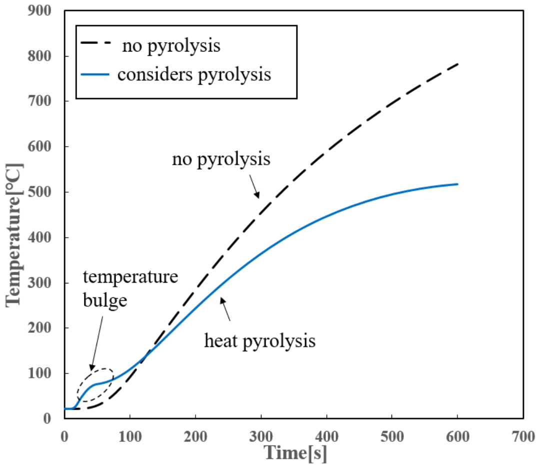
(a) the influence of contact thermal resistance on outer wall temperature



(b) the influence of thermal conductivity of insulation layer on outer wall temperature



(c) the influence of extra heat source on the temperature change of outer wall surface



(d) the influence of pyrolysis effects on the outer wall temperature against operation time

Figure 10 Variation of metal outer wall temperature with time

Figure 10 present the effects of influence factors on the variation of metal outer wall temperature against operation time. The figure 9(a) shows the addition of interlayer thermal conductivity leads to a significant decrease of the wall temperature from 100s, furthermore, the wall temperature decreases with the decrease of the value of interlayer thermal conductivity. In figure 9(b), a similar trend appears, i.e., the wall temperature decreases with the decrease of thermal conductivity of insulation layer. However, the effects mostly take place at the region after 300s, where the temperature rises up to 500℃. In a short, the above two factors are conductive to reduce the maximum wall temperature. Two phenomenons can be found in figure 9(c), one is the temperature bulge at the very first stage, i.e., before 100s, it is attribute to the pyrolysis gas penetrate and flows to the metal shell layer, brings an extra heat flux leading to the sharp increase of wall temperature. The other one is the tightening of the wall temperature profile, which is caused by a negative heat source generated during the pyrolysis of carbon phenolic. The size of temperature bulge or temperature tighten depends on the magnitude of heat flux of pyrolysis gas or that of heat absorption. The figure 9(d) presents a curve considers all the effects brought by the pyrolysis of carbon phenolic, comparing with the other curve with no pyrolysis. The results present above provide a reference to the design or fault detect of MPTPS in combustion with pyrolytic materials in combustion.

CONCLUSION.

In this paper, a one-dimensional transient heat conduction model has been constructed to understand the thermal insulation procedure of multilayer passive thermal protection system(MPTPS). The model has been build by adopting the finite element method, considering the effects brought by the pyrolysis of carbon phenolic composite, include thermal resistance of interlayer gap, the variation of thermal properties of thermal insulation layer and the extra heat source. In order to investigate the pyrolysis characteristics of carbon/phenolic composites, some experimental studies have been carried out. The conclusions are summerized as follow:

1) The main pyrolysis products of carbon phenolic at the temperature range of 400-700℃are carbon dioxide, phenol and its derivative, whereas after 800℃， the carbon dioxide will be dominant.

2) The pyrolysis of carbon phenolic composite is mainly the pyrolysis of phenolic resin material, which completely pyrolyzed after some time. According to the present results, the mass of sample decreases rapidly at the temperature range of 400-600℃, while the mass remains unchanged after 700 700℃. The sample keeps a complete surface morphology from the view of its scanning pictures.

3) Both the addition of thermal resistance of interlayer gap and the decrease of thermal conductivity of insulation material are conductive to reduce the maximum metal wall temperature.

4) The pyrolysis of carbon/phenolic materials has great influence on the temperature change of the outer wall, such as the temperature bulge results from the penetration of gas, the temperature curve tightening results from the heat absorption. The size of temperature bulge or temperature tighten depends on the magnitude of heat flux of pyrolysis gas or that of heat absorption.

NOMENCLATURE

ε	porosity
k_{eff}	equivalent thermal conductivity, W/m-K
k_f	thermal conductivities of the air, W/m-K
k_s	thermal conductivities of the solid, W/m-K
Φ	heat absorbtion of pyrolysis,w/m^3
q	heat flux of pyrolysis gas flows to the outer metal wall,w/m^2
t	Time,s

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