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Effects of surface temperature, flow velocity and ambient pressure on the oxidation of graphite at 1400–2000 °C in O_2

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ABSTRACT

The effects of surface temperature (1400–2000 °C), flow velocity gradient (130–600 s^{-1}) and ambient pressure (5–101 kPa) on the oxidation behavior of graphite were studied in O_2. The weight loss rate between 1400 and 1800 °C is independent of the surface temperature but slightly decreases at 2000 °C. Besides, it displays quasi-parabolic increases with increasing the flow velocity gradient and the ambient pressure but levels off above the ambient pressure of 20 kPa. According to the theoretical analysis, the enhancement of the gas-phase CO–O_2 reaction has a joint reduction effect on the weight loss rate.

1. Introduction

Recently, evaluation of the high temperature oxidation/ablation resistance of various graphite-based [1–5] and C/C-based [6–11] composite materials is of great interest due to the demand of advanced thermal protection systems (TPS) for hypersonic vehicles. The typical environment related to hypersonic flight involves oxidizing atmospheres with temperatures above 1400 °C and dynamic pressures in the range of 100–2000 psf (~5–96 kPa) [12,13]. In order to understand clearly the oxidation/ablation mechanism of these composite materials in the above extreme environments, oxidation behavior of graphite as the base material under various conditions needs to be well understood in the first place.

High temperature oxidation of graphite and C/C has been subject to an extensive amount of experimental and theoretical investigations due to its practical importance in aerospace industry [14–22], nuclear energy industry [23,24] as well as in coal/char combustion industry [25]. Usually, the stagnation gas flow configuration is employed in the investigation. This flow filed is well-defined and can be characterized uniquely by a single parameter, the velocity gradient *a*. In this way, not only does it well simulates the situations in practical applications but also facilitates mathematical analysis and interpretation of the oxidation behavior of graphite.

It is generally known that, at high surface temperatures ($\gtrsim$ 1116 °C in [14,18], $\gtrsim$ 800 °C in [24], $\gtrsim$ 1027 °C in [26]), the oxidation of graphite becomes boundary layer diffusion-controlled. In this diffusion-controlled regime, the weight loss rate of graphite depends weakly on the surface temperatures [25] but strongly on the boundary layer characteristics. In high velocity gas flows, the boundary layer characteristics are mainly determined by the composition, pressure and velocity gradient [27,28]. Scala [14] has theoretically established a boundary layer model for the oxidation of graphite in hypersonic airflows. It was expressed that the weight loss rate of graphite in the diffusion-controlled regime is proportional to the half power of the surface stagnation pressure. Subsequently, Metzger et al. [17] investigated the oxidation and sublimation of graphite in arc-jets with the surface temperature from 1200 K (~927 °C) to ~4000 K (~3727 °C). They compared their experimental results with the theoretical solutions based on the theories proposed by Scala and Gilbert. Close agreement was announced after correlation. Matsui et al. [26] investigated the dependence of the combustion rate of solid carbon on the flow velocity at surface temperatures of 1200–1700 K (~927–1427 °C). Similarly, it was concluded that the combustion rate is proportional to the half power of the flow velocity gradient for surface temperature above 1500 K (~1227 °C). However, in relatively low velocity flows, the change of surface temperature and the parameters of the oxidizing gas flow mentioned above may also affect the appearance or the intensity of the gas-phase CO–O_2 reaction inside the boundary layer, which can cause joint influences on the boundary layer characteristics and thus the weight loss rate of graphite. For that, Adomeit et al. [29] theoretically analyzed the experimental results obtained by Matsui et al. [26] and proposed a boundary layer model for the combustion of solid carbon in an axisymmetric, quasi-steady, and laminar stagnation gas flow field. Finite gas-phase reaction rates as well as limiting cases of frozen flow and an infinitely fast gas-phase reaction were taken into

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Nomenclature

a	flow velocity gradient, in s^{-1}
A_{si} ($i = O, P$)	reduced surface Damköhler number for C–O_2 surface reaction (R1) and C–CO_2 surface reaction (R3), respectively
B_{si} ($i = O, P$)	frequency factor for C–O_2 surface reaction (R1) and C–CO_2 surface reaction (R3), respectively, in m/s
E_{si} ($i = O, P$)	activation energy for C–O_2 surface reaction (R1) and C–CO_2 surface reaction (R3), respectively, in kJ/mol
K	factor
$\dot{w}$	weight loss rate, in kg/m^2 s
P_∞	ambient pressure, in kPa
T_∞	ambient temperature, in K
T_s	surface temperature, in K
W_i ($i = C, O, P$)	molecular weight of C, O_2, CO_2, respectively, in kg/mol
β	mass transfer number
μ_∞	flow viscosity in the free stream, in Pa s
ρ_∞	flow density in the free stream, in kg/m^3

account in their calculations. It was mentioned that the combustion rate was strongly influenced by the gas-phase reaction rate at about 1200–2000 K (~927–1727 °C). Makino and coworkers [30–33] further experimentally investigated the combustion of a graphite rod in a two dimensional stagnation gas flow above 1000 K (~727 °C) and theoretically extended the chemically reacting boundary layer model. To our best knowledge, previous investigations have mainly focused on the temperature dependence of the oxidation/combustion behavior of graphite, but little work has been done specifically towards the effects of flow velocity gradient and ambient pressure on the appearance of the gas-phase reaction and then their joint influences on the oxidation rate of graphite at above 1400 °C in the boundary layer diffusion-controlled regime, which is of great practical importance to hypersonic applications.

In the present work, first, we studied the oxidation behavior of graphite in the temperature range of 1400–2000 °C. And then, the effects of flow velocity gradient (130–600 s^{-1}) and ambient pressure (5–101 kPa) on the oxidation behavior of graphite at 1600 °C were investigated. The obtained experimental results were compared with the chemically reacting boundary layer model proposed by Makino et al. [25]. According to that, the effects of flow velocity gradient and ambient pressure on the appearance of the gas-phase CO–O_2 reaction and their joint effect on the weight loss rate of graphite were analyzed, which can provide useful information for better understanding the oxidation/ablation mechanism of the graphite-based and C/C-based composite materials in related extreme environments.

2. Experimental

2.1. Material

A commercial graphite, named as MCL-82 (Shanghai New Carbon, Co., Ltd.), was used in this work. The main properties of this graphite are listed in Table 1. The samples for the oxidation testing were cut and machined into cylinders of 20 mm in diameter in order to produce low velocity gradients.

Table 1
Main properties of MCL-82 graphite.

Property	MCL-82
Density (g/cm^3)	1.82
Porosity (%)	19.5
Compression strength (MPa)	~60
Flexural strength (MPa)	~33
Electrical resistivity (μΩ m)	~15
Ash rate (ppm)	<50

2.2. Oxidation testing setup

An induction heating oxidation testing apparatus has been established in our laboratory. Its configuration is schematically shown in Fig. 1. The graphite cylinder sample was inductively heated. The surface temperature of the sample was measured by a two color ratio pyrometer (Heitronics KT 18.03 R, 1000–3000 °C). Dry O_2 or air from the commercial gas cylinder was used as an oxidizer. The oxidizing flow entered the testing chamber through the nozzle with a uniform velocity, and passed over the sample to establish a two-dimensional stagnation flow. The flow field was characterized uniquely by the velocity gradient ($a = 4V/d$), where V is the free stream velocity and d is the diameter of the graphite cylinder. To measure the oxidation rate, a micro-vision camera was installed to monitor and photographically record the dimensional changes of the sample during each test at a resolution of ±0.02 mm and at an interval of 1 s.

2.3. Oxidation testing procedure

During each oxidation test, the testing chamber was evacuated to a pressure of ~2 Pa prior to heating. Then, the sample was heated at an average heating rate of 20 °C/s to the target temperature. After a 5 min isothermal hold, dry O_2 or air was rapidly introduced into the testing chamber through a leak valve. Normally, the desired gas pressure in the testing chamber was

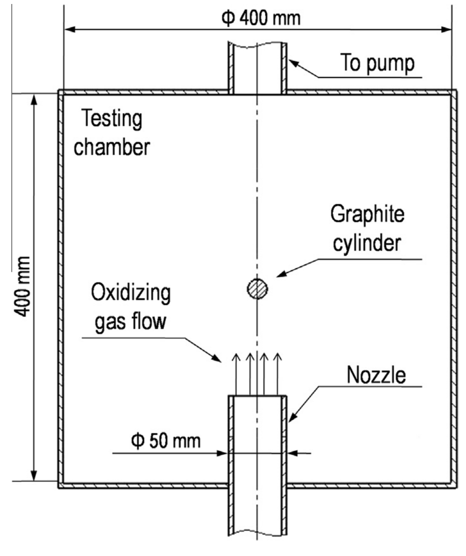

Fig. 1. Schematic of the induction heating oxidation testing apparatus.