Oxidation behavior of a multilayered (Si-B-C) ceramic in a complex atmosphere $N_2/O_2/H_2O$

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INTRODUCTION

A (Si-B-C) matrix, constituted of different layers as silicon carbide (SiC), boron carbide (B_4C) and Si-B-C phase, has been developed to improve the oxidation resistance and lifetime in an oxidizing environment of SiC/SiC composites. The incorporation of boron compounds in the matrix allows the formation of boron oxide and borosilicate during oxidation at a lower temperature than that of the silica formation ($T \approx 1000°C$). In addition, the glassy SiO_2/B_2O_3 phase may flow along the matrix cracks, created when the elastic limit is exceeded during a mechanical solicitation. These oxides formed by the oxidation of the different layers lead to the crack sealing, stopping the oxidizing agents diffusion (e.g. O_2 and H_2O) to the fibers by gas–phase diffusion [Fig. 1].

The aim of this work is to estimate the rates of the oxide formation (e.g B_2O_3 and borosilicate) and the carbide recession (e.g B_4C and Si-B-C phase) at atmospheric pressure, as function of temperature $T \in [600°C-1400°C]$ and O_2 and H_2O partial pressures ($P_{O_2}=[0-20kPa]$ and $P_{H_2O}=[0-15kPa]$) for each kind of coatings. The mass variation per surface unit ($\Delta m/S$) versus time for boron-containing coatings, was measured with a thermogravimetric analysis apparatus (SETARAM Setsys 16/18, France), to evaluate both the oxide formation and recession rates.

This approach should allow to quantify the self-healing of a multilayered matrix and to understand the different oxidation mechanisms, taking into account the specificity of the boron compounds (e.g the B_2O_3 volatilization, its water sensibility and the B_2O_3-SiO_2 flowing) .

Results and discussion

The oxidation rate of flat B_4C coatings under dry atmosphere, in a domain of temperature ranging from 600°C to 900°C, is correctly described by a parabolic law [Fig. 2 a)], indicating that the growth of the oxide layer is limited by the diffusion of dioxygen. Above 1000°C, the B_2O_3 volatilization and flowing have to be considered , as those two phenomena make more complex the interpretation of thermogravimetric curves.

Under wet atmosphere, there is a competition between the growth rate (O$_2$ effect) and volatilization rate (H$_2$O effect) of B_2O_3. When both rates are similar, a paralinear model is applicable [Fig. 2 b)] [1]. On the other hand, if the volatilization rate is higher, oxidation regime becomes active [Fig. 2 c)].

The oxidation rate of flat Si-B-C coatings is more complex, because of the evolution of the borosilicate composition versus time and temperature. The effect of the aging of the borosilicate in different oxidizing environments, resulting from the B_2O_3 volatilization during time, has also been investigated.

This analysis is useful to explain the different behaviors of the Si-B-C matrix in an oxidizing environment. Under dry atmosphere (T<900°C), the self-healing of the Si-B-C matrix is mainly achieved by B_2O_3. Under wet atmosphere (T=600°C), it is obtained by Si-rich borosilicate due to the oxidation of the Si-B-C coatings. Thus, B_2O_3 alone as sealing is not fully efficient because of its high reactivity with water. Finally, the B_2O_3 volatilization rate along the matrix crack is simulated and compared with that determined owing to the weight loss of B_2O_3 (raw material) determined versus time by thermogravimetric analysis.

ACKNOWLEDGEMENT

REFERENCES

1. E.J. Opila, J. Am. Ceram. Soc., **80**, 197 (1997)

Fig. 1 : Schematic of the crack opening under mechanical loading and the sealing process in multilayered ceramic matrix composites
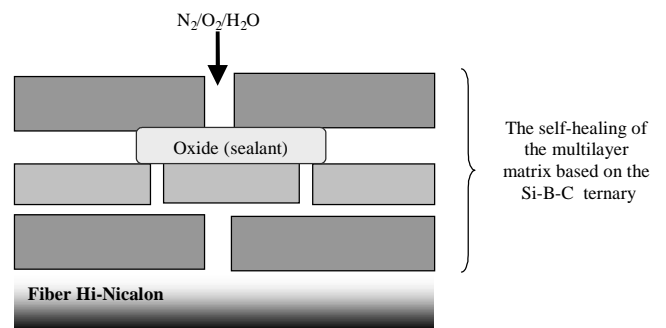
Fig. 2 : Different oxidation regimes of flat B_4C coatings in an oxidizing environment ($N_2/O_2/H_2O$)

a) Parabolic regime: 800°C N_2/O_2 (80/20)
b) Paralinear regime: 800°C $N_2/O_2/H_2O$ (70/20/10)
c) Active regime : 600°C $N_2/O_2/H_2O$ (70/20/10)

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