

MECHANICAL AND ELECTRICAL PROPERTIES OF POLYSILOXANE-MODIFIED CARBON-CERAMIC COMPOSITES

T. Gumula*, D. Mis

AGH University of Science and Technology, Faculty of Materials Science and Ceramics, Al.
Mickiewicza 30, 30-059 Krakow, Poland

*tgumula@agh.edu.pl

Keywords: carbon-ceramic composites, polymeric precursors, polysiloxanes, carbon fibres

Abstract

The aim of this work was to investigate properties of inexpensive multifunctional carbon-ceramic composites, which future application will be load bearing heating elements. The carbon-ceramic composites were obtained by impregnation of C/C composites with polysiloxane preceram, followed by heat treatment at 1000°C in an inert atmosphere. Mechanical and electrical properties of carbon-ceramic composites were investigated.

The results indicate that incorporation of polysiloxane-derived ceramic phases into C/C composites does not have significant influence on their mechanical properties.

The temperature resistance coefficient values for C/C and carbon-ceramic composites are similar and indicate that they behaved as linear resistors. Carbon-ceramic composites reinforced with UTS fibres can be used as heating elements with watt loading similar to C/C composites, namely up to about 80-100 W/cm².

1. Introduction

The rapidly developing materials science seeks new multifunctional materials that can work at high temperature and in reactive and changing atmospheres.

Carbon-carbon (C/C) composites are candidates for high-temperature applications. They possess appropriate values of thermal conductivity and electrical resistivity [1,2]. Moreover, C/C composites are light, retain their high strength and stiffness at high temperatures (up to 2000°C), and due to low CTE and a high heat of sublimation they have good ablation resistance [1,2]. The main disadvantage of carbon-based materials is their low oxidation resistance [1,2]. For this reason, ceramic materials characterised by good oxidation resistance have been progressively incorporated in the carbon-based materials to protect them against the environment.

The aim of this work was to investigate mechanical and electrical properties of inexpensive multifunctional carbon-ceramic composites, which future application will be load bearing heating elements. Similarly to C/C composites, they should retain high strength and modulus at high temperature, however, due to presence of ceramic phase, they should perform distinctly higher oxidation resistance. Moreover, their fracture toughness should be higher than this for silicon carbide and molybdenum disilicide, which are typical materials that heating elements are made from.

The composites were obtained by impregnation of C/C composites with polysiloxane preceram, followed by heat treatment at 1000°C in an inert atmosphere. Using the preceram is

a new, very promising way to obtain carbon-ceramic composites and to protect them against oxidation [4]. Structure, microstructure and properties of ceramic phase depends on polymer precursor and processing parameters [3,4].

2. Materials and testing methods

The C/C composites were manufactured by the liquid impregnation procedure. As reinforcing material 2D carbon fabric and carbon roving were used (see Table 1). Matrices of C/C composites were obtained from the mixture of Novolac MR phenol-formaldehyde resin (ZCH Organika – Sarzyna, Poland) with UF15 SiC powder (H. C. Starck StarCeram S; grains diameter P 50% = 0.55 μ m). In order to obtain carbon-ceramic composites, C/C composites were impregnated with commercially available polysiloxane-based solution of preceram mixed with mentioned above SiC powder and subjected to heat treatment up to 1000°C in an inert atmosphere. Lukosil 901 polymethylphenylsiloxane resin, produced by Lucebni zavody, Kolin (Czech Republic) was used as preceram. Types of prepared samples are shown in Table 1. Porosity of reference samples was 20 \pm 3% and density 1.37 \pm 0.07 g/cm³, while porosity of densified samples was 12 \pm 2% and density 1.50 \pm 0.01 g/cm³. Densified composites (samples 7-12) were obtained from reference composites (samples 1-6), respectively (see Table 1).

Labelling	Description	Carbon fibres	Matrix – phase I	Matrix – phase II
Reference samples (C/C composites and C/C composites with their matrix modified with SiC powder)				
1	Sample 1	2D fabric, HTS 5131 196 g/cm ²	C	-
2	Sample 2	2D fabric, HTS 5131 196 g/cm ²	C-SiC	-
3	Sample 3	Roving STS 5631	C	-
4	Sample 4	Roving STS 5631	C-SiC	-
5	Sample 5	Roving UTS 50 F13	C	-
6	Sample 6	Roving UTS 50 F13	C-SiC	-
Densified samples (carbon-ceramic composites)				
7	Sample 1, densified	2D fabric, HTS 5131 196 g/cm ²	C	SiCO-SiC
8	Sample 2, densified	2D fabric, HTS 5131 196 g/cm ²	C-SiC	SiCO-SiC
9	Sample 3, densified	Roving STS 5631	C	SiCO-SiC
10	Sample 4, densified	Roving STS 5631	C-SiC	SiCO-SiC
11	Sample 5, densified	Roving UTS 50 F13	C	SiCO-SiC
12	Sample 6, densified	Roving UTS 50 F13	C-SiC	SiCO-SiC

Table 1. Types of prepared composites

The strength and Young's modulus the composite samples were determined in three point bending test on Zwick machine model –1435.

Electrical measurements were conducted in quartz tube in an inert atmosphere (argon). Samples in a form of bars were placed in graphite grips, achieving electrical contact. The current – voltage relationships were registered. The temperature changes of the composites accompanying electrical parameters changes were monitored by means of optical pyrometer in the range from 800°C to 1500°C.

The resistance of composite samples was calculated from the Ohm's law, and coefficient of temperature resistance was calculated from the formula:

$$CTR = \Delta R / (R_0 \Delta T)$$

where: CTR - coefficient of temperature resistance [1/K]

ΔR – change in resistance due to temperature change [Ω]

ΔT – temperature difference [$^{\circ}C$]

R_0 – resistance measured at room temperature [Ω]

The quantity of dissipated electrical power on composite samples (watt loading) was calculated from the formula:

$$P_s = P/S$$

where:

P – power [W]

P_s – watt loading [W/cm^2]

S – composite surface [cm^2]

3. Results

Figure 1 shows the relative change of mechanical properties for densified composites (samples 7-12) in relation to reference composites (samples 1-6). As it indicates from this diagrams densification process do not have influence on bending strength of the composites. Small influence of densification process on Young's modulus values can be visible. Young's modulus has increased about 15% in case of samples 7-10. It can be associated with an increase of the amount of ceramic phase in composites. Densification leads to decrease of elongation to break. Densified carbon-ceramic composites (samples 7-12) have 30% lower elongation than the reference composites (samples 1-6).

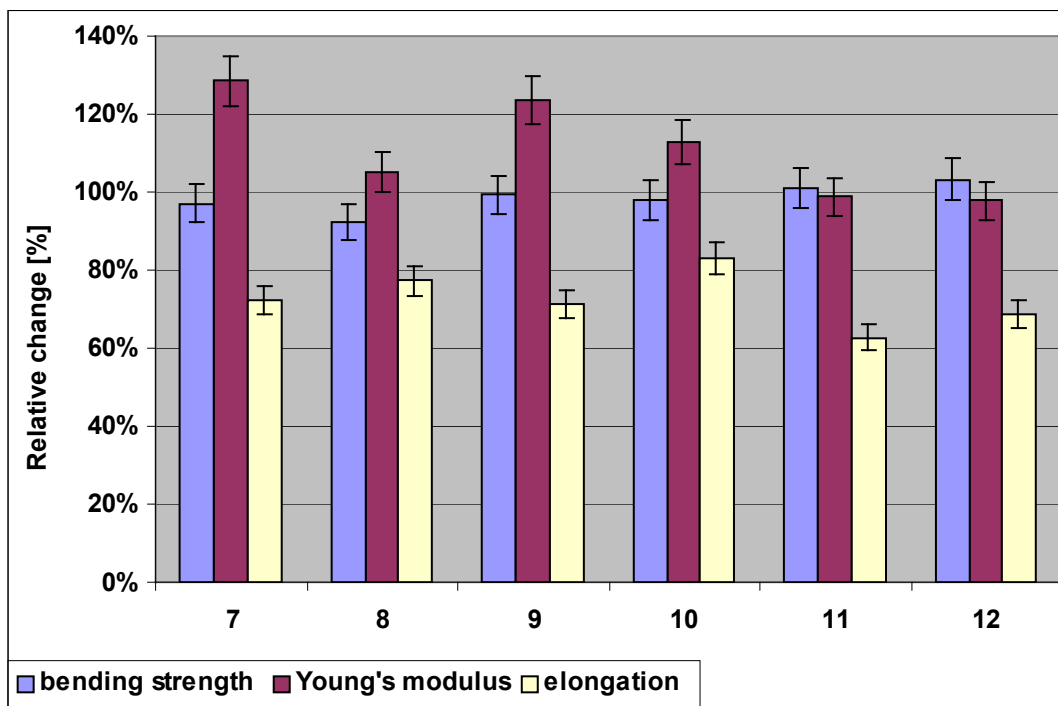


Figure 1. Relative change of mechanical properties for densified samples in relation to their reference samples

Watt loadings of ceramic matrix composites calculated at 1500°C are given in Figure 2. In general, watt loading on a heating element and temperature limit of application are the most important factors in the design of heating elements. The element temperature is dependent on how much energy has to be transmitted from the heating element to the surrounding area, the temperature of the surrounding area, and on heat transfer from the element to the surrounding area. Generally, the higher watt loading capability is advantageous parameter of a material for heating element. Reducing of watt loading requires to use larger heating element and an increase of costs.

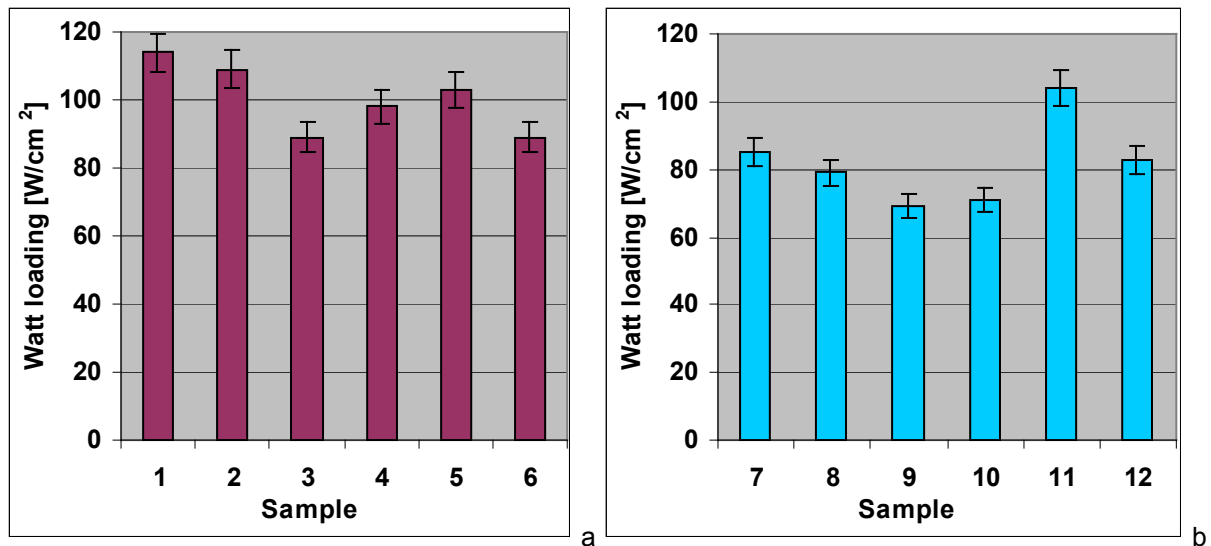


Figure 2. Watt loading of a) reference samples, b) densified samples

The practical limit of watt loadings for conventional heating resistors are as it follows: metallic – from 8 to 12 W/cm², silicon carbide – from 10 to 15 W/cm², molybdenum disilicide – from 20 to 30 W/cm² [5]. As it results from Figure 2, the watt loading values of investigated composites are distinctly higher in comparison with conventional heating resistors.

Densification procedure results in decrease of the watt loading of composites. In case of samples 7-10 the parameter decreases by about 25 %, while comparing them with reference samples 1, 2, 3 and 4 (Figure 2). Only composite samples reinforced with UTS fibres, namely samples 11 and 12, do not exhibit losses of watt loading after densification, in relation to reference samples 5 and 6.

The coefficient of thermal resistance were calculated from the current – voltage dependence in the temperature range from 800°C to 1500°C. The calculated CTR values for both composites are similar and low. The CTR values, ranges from $-3 \cdot 10^{-4}$ [1/°C] to $-5 \cdot 10^{-4}$ [1/°C], indicate that all composites (reference and densified) behave as linear resistors and densification has no influence on this parameter. Negative values of this parameter suggest that electrical conductivity of composites is controlled by electrical conductivity of carbon fibres (semiconductors).

4. Conclusions

The results indicate that incorporation of polysiloxane-derived ceramic phases into C/C composites does not have significant influence on their bending strength and Young's modulus. Carbon-ceramic composites does not exhibit brittle behaviour typical for ceramics.

The temperature resistance coefficient values for C/C and carbon-ceramic composites are similar and indicate that they behaved as linear resistors. Carbon-ceramic composites reinforced with UTS fibres can be used as heating elements with watt loading similar to C/C

composites reinforced with UTS fibres, namely up to about 80-100 W/cm². However, their possible use as heating elements requires further investigation on long term reliability, under thermal shock and in oxygen-containing atmosphere conditions.

Acknowledgements

This work was supported by budgetary resources of the Polish Ministry of Science and Higher Education in years 2012-2014, grant Iuventus Plus No IP2011 016771.

References

- [1] Burchell T. D. Carbon Materials for Advanced Technologies. Pergamon, 1999
- [2] Savage G. Carbon – carbon composites. Chapman and Hall, London, 1992
- [3] Gumula T., Michalowski J., Blazewicz M., Blazewicz S. A microstructural study of carbon-carbon composites impregnated with SiC nanofilaments. *Ceramics International*, **36**, pp. 749-753 (2010)
- [4] Gumula T., Paluszkiwicz C., Blazewicz S. Study on thermal decomposition processes of polysiloxane polymers – from polymer to nanosized silicon carbide. *Journal of Analytical and Applied Pyrolysis*, **86**, pp. 375-380 (2009)
- [5] Kanthal Handbook. Resistance Heating Alloys and Systems for Industrial Furnaces. Sweden, 2001