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A hybrid ceramic-polymer composite fabricated by co-curing lay-up process for a strong bonding and enhanced transient thermal protection

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ABSTRACT

A hybrid ceramic-polymer composite is fabricated by a co-curing lay-up process by combining a carbon nanotube (CNT) reinforced ceramic composite thin film with a carbon fiber reinforced polymer (CFRP) composite substrate. The ceramic nanocomposite thin film has good flexibility, thermal conductivity and high temperature tolerance. The polymer composite substrate is a carbon fiber reinforced bismaleimide composite that is widely used in aerospace and automotive industries. Finite element analysis (FEA) is used to investigate the maximum survival temperature with different thicknesses of the ceramic nanocomposite. The resultant hybrid composite shows good structural integrity and displays a pull-off bonding strength up to 8.3 MPa. In addition, thermal study illustrates that such a flexible CNT reinforced ceramic composite can effectively protect CFRP in an elevated temperature environment by delaying transient thermal conduction.

1. Introduction

Carbon fiber reinforced polymer (CFRP) composites (also known as polymer matrix composites (PMC)) are a type of strong and light weight composite material, that are commonly used in aerospace, automotive and civil engineering applications [1–4]. Modern airplane structures are commonly made up of 20–70% of such fiber reinforced composite materials [5]. For example, Boeing 787 aircraft is designed with CFRP for fuselage, wings, and other key airframe components. However, the material has inherent drawbacks that limit its viability under high temperature conditions. The most common matrix materials for CFRP composites are epoxy and bismaleimides (BMI), whose maximum service temperatures are about 190 °C and 232 °C [6], respectively. When used in thermal critical areas such as leading edges of supersonic vehicles or turbine fans of aero engines, the polymer matrix will not perform as desired. This is due to the thermal softening effect – strength and modulus degradation at high temperatures. Additionally, cracks and fractures may develop after long periods of exposure to extreme temperatures [7–9].

Extensive research has been done to improve the thermal tolerance of CFRP composites. Different kinds of thermal barrier coatings have been developed to protect CFRP from harsh thermal environments [10–15]. These coatings are mostly metal-based and used in order to build a good bonding with the CFRP substrate. It is time and energy consuming to establish an effective coating on CFRP. Firstly, surface pretreatment is always required because cured resin on CFRP surface is inert for mechanical and chemical bonding. Then, multi-layer structure with different composition in each layer is essential to reduce the thermal expansion mismatch between metallic material and CFRP. In addition, metals possess high isotropic thermal conductivity properties, which makes protecting the CFRP from thermal impact and overheating extremely difficult, especially for aerospace structural materials. Ceramic coatings have outstanding thermal insulation properties. However, it is difficult to develop a ceramic coating in-situ with the CFRP manufacturing process due to the large difference between the required high temperatures during preparation. Direct adhesion of ceramic material on CFRP results in delamination because of the low bonding strength. Ceramic coatings are usually applied by a spraying method, which projects ceramic particles onto the CFRP surface and embeds the particles into the deformed polymer matrix [16,17]. As a result, the surface morphology is poor and coating is not continuous, which degrade the mechanical strength of the CFRP substrate and deteriorate the thermal barrier performance.

This research presents a new method of developing a hybrid composite material to improve transient thermal protection of traditional CFRP composites [18,19]. By using a standard co-curing lay-up process

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for polymer composites, in which a ceramic thin film is bonded onto the polymer composites to ensure the material's integrity, a new and effective hybrid composite can be manufactured. Finite element modeling is performed to study the maximum heat source temperature with specific ceramic film thickness in order to avoid the glass transition of resin in CFRP substrate. Influence of different ratio of ceramic to polymeric in matrices of the transition layer on the bonding strength is investigated. The bonding strength and failure mechanism correlated with the fabrication conditions are discussed. The surface morphologies of the hybrid composite structure are characterized by scanning electron microscopy (SEM). Transient thermal protection effect is also tested experimentally.

2. Materials preparation

2.1. Materials

Random carbon nanotube (CNT) sheets (Nanocomp Technologies, Inc., Merrimack, NH, US), comprised of multi-walled carbon nanotubes (diameter as 6–8 nm and length around 1 mm), were used as the preforms for the ceramic nanocomposite. Polysilazane (KiON Defense Technologies, Inc., Huntingdon Valley, PA, US), a liquid thermosetting resin with repeating units in which silicon and nitrogen atoms are bonded in an alternating sequence, was used as the liquid preceramic precursor to prepare the CNT reinforced ceramic matrix nanocomposites. Non-woven carbon fiber sheets (ACP Composites, Inc., Livermore, CA, US) with an area density of 0.5 oz/yard^2, were used as the transition layer material. 375 carbon fiber/5250-4 Bismaleimide (BMI) equivalent prepreg (Stratton Composite Solutions, Marietta, GA, US) with ply thickness around 1 mm was used to prepare the CFRP composites.

2.2. Preparation of CNT reinforced ceramic composite and the transition layer

CNT reinforced ceramic matrix nanocomposites were produced through a process known as polymer impregnation and pyrolysis (PIP) [20,21]. The preform has an average pore size of 15.4 nm and porosity percentage of 71.8%. Here, the CNT preform was immersed into polysilazane (liquid state, ceramic precursor) and then subjected under vacuum (Step (1) in Fig. 1). After taking them out from the polysilazane bath, the residual liquid was wiped off and a controlled volume of the precursor drops were applied on the top surface (Step (2) in Fig. 1). Multi-layer non-woven carbon fiber tissues were stacked on top to create the transition layer (Step (3) in Fig. 1). This is designed to enhance the bonding strength and minimize the coefficient of thermal expansion (CTE) mismatch between the ceramic nanocomposite and the CFRP layer. The volume of precursor drops (Step (2) in Fig. 1) is one important factor, which affects the bonding strength between the ceramic nanocomposite and CFRP substrate. Such liquid precursor drops infiltrate into the transition layer. The amount determines the ratio of ceramic matrix to the polymer matrix and decides the interface area and interlocking density. The interlocking effect will directly influence the bonding strength. Detailed fabrication procedure and measurement results are explained in Section 3.2. Then, the stack of sample was cured into a solid piece by thermal crosslinking at 140 °C for 24 h under pressure in air atmosphere (Step (4) in Fig. 1), and then were through pyrolytic transformation at 1000 °C for 1 h in nitrogen atmosphere (Step (5) in Fig. 1).

2.3. Preparation of hybrid ceramic-polymer composite

Pores/voids increase in the transition layer after pyrolysis due to the volume shrinkage during polymeric precursor to ceramic conversion. Based on the experimental results, extra resin in carbon fiber reinforced BMI prepreg cannot fill the pores in the transition layer. Therefore, as shown in step (6) in Fig. 1, extra BMI/acetone solution was used to infiltrate the transition layer. Such infiltration process (Step (7) in Fig. 1) was repeated three times in order to fully fill the pores in the transition layer. In Step (8) in Fig. 1, three layers of carbon fiber reinforced BMI prepreg were laid up on top of the transition layer to form a laminate structure. The laminate structure was sealed in a vacuum bag and was co-cured (Step (9) in Fig. 1).

2.4. Thermal property measurement setup

Thermal gravimetric analysis (TGA) is processed on TA Q50 (TA instruments, Inc., New Castle, DE, US) under air atmosphere using 10 °C/min heating rate. Fig. 2 is the experimental setup to measure the transient thermal protection effect of the CNT ceramic composite. Three layers of CNT ceramic composites (~ 60 µm) were used as coating to achieve better thermal protection. In order to ensure a good contact between the thermocouple and the sample, as well as the sample to the hot plate, double-sided carbon tape was applied to hold the two surfaces together. A CFRP composite and a hybrid composite were placed at the same position on the hot plate, where for the latter sample, the ceramic thin layer is laid facing downward to the hot plate to protect from the heat generated. The hot plate temperature was set at 120 °C.

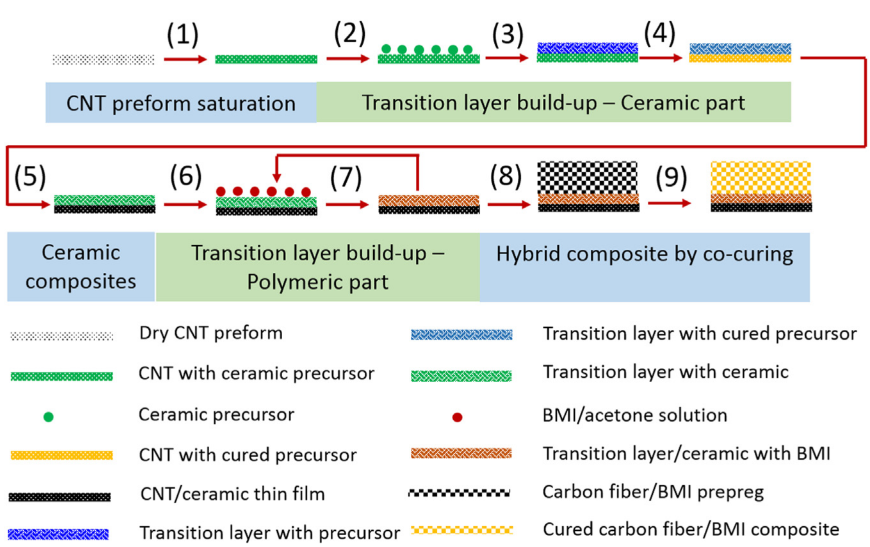

Fig. 1. Co-curing lay-up procedure to bond a ceramic composite thin layer on the outside surface of CFRP composites.