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Effect of rice-husk ash on properties of laminated and functionally graded Al/SiC composites by one-step pressureless infiltration

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ABSTRACT

The quantitative effect of the following parameters on the one single step pressureless infiltration characteristics of bilayer SiC_p/rice-husk ash (RHA) porous preforms by aluminum alloys was investigated using the Taguchi method and analysis of variance (ANOVA): infiltration temperature and time, SiC particle size, RHA percentage, percentage porosity in the preforms, and magnesium content in the alloy. The contributions of each of the parameters and their interactions to the retained porosity, hardness and modulus of elasticity of the resulting bilayer composites were determined. The parameters that most significantly impact the modulus of elasticity (E) of the resulting composites are infiltration time and SiO_2 phase type (from RHA), with contribution of 27% and 21%, respectively. They are followed by preform porosity, Mg concentration and process temperature, with contributions of 13%, 12% and 11%, correspondingly. Verification tests conducted using the established optimum parameters show a good agreement with the projected values of modulus of elasticity (169 ± 9 GPa) and retained porosity (1.9 ± 0.8). The in-situ formation of $MgAl_2O_4$ and MgO phases with the unique morphology of RHA in the microstructure, was manifested as exothermic peaks in the heat flow curves and identified by XRD and SEM analyses of the composites. A reaction pathway for $MgAl_2O_4$ and MgO formation was outlined.

1. Introduction

Functionally graded composite materials (FGCMs) have been the subject of intense research efforts in recent years. Several studies have examined the spatial variation of mechanical properties to optimize structural response. Others have considered processing techniques which produce structures with variable reinforcement and thus variable mechanical properties [1–3]. The manufacturing process of a FGM can usually be divided into building the spatially inhomogeneous structure "gradation" and transformation of this structure into a bulk material "consolidation". Gradation processes can be classified into constitutive, homogenizing and segregating processes. Constitutive processes are based on a stepwise build-up of the graded structure from precursor materials or powders. In homogenizing process a sharp interface between two materials is converted into a gradient by material transport. Segregating processes start with a macroscopically homogeneous material which is converted into a graded material by material transport caused by an external filed. Homogenizing and segregating processes produce continuous gradients, but have limitations concerning the types of gradients which can be produced [3].

Although the infiltration of ceramic preforms by liquid metals has been typically applied for the processing of composite materials with homogeneous reinforcement shape, size and composition [4,5], as a constitutive route, it offers the potential for the production of graded materials by variation of shape, size, and volume fraction of the reinforcement in superimposed ceramic perform layers over a metallic layer.

In this investigation, it is suggested that such graded materials may be produced by one-step infiltration process. Thus, components such as cylinder liners, brake components and ballistic armor plates may be produced by adequately packing two or more ceramic preform layers of distinct characteristics. Nonetheless, a number of difficulties related to the processing parameters (infiltration time and temperature, process atmosphere, alloy composition, shape and size of the reinforcement, and percentage porosity in the preform, etc.) are encountered in the production of MMCs via the infiltration of ceramic preforms, particularly when using the capillary effect. In the Al/SiC system, for instance, typical problems are the presence of aluminum carbides and residual porosity in the composites [6]. In spite of various efforts devoted to the

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optimization of wettability and pressureless infiltration parameters [4,7], it is expected that in pressureless infiltration-based processing, changes of key parameters (reinforcement composition, size, percentage porosity in the preform) from one section to another within the ceramic preform (layer by layer) will increase the inherent barriers and challenges. It was reported that some of the problems associated with the infiltration process can be solved by addition special elements or phases [8]. Furthermore, addition of reinforcement phases with special morphology and chemical composition can enhance mechanical and physical properties of the composites. Rice husk ash (RHA) as a source of silicon and oxygen with truss like and spiral like structure can be used as a unique reinforcement phase to satisfy these expectations [9]. However, the past and current fabrication methods do not allow exploiting the RHA reinforcementś full potential because during processing the natural structure of the ash can be broken easily. For instance, stir casting and powder metallurgy routes may lead to the rupture of the truss like and spiral like structure of the as-burned ash [10–14]. The aim of this investigation is to gain further insight into the processing of functionally graded Al/RHA/SiC_p composites by pressureless infiltration. In this article, the quantitative effect of process temperature and time, SiC_p particle size in the preform layer, crystallinity of SiO_2 obtained from rice husk ash, ceramic preform porosity and Mg concentration in the aluminum alloys on the infiltration characteristics of RHA/SiC_p ceramic preform layers is investigated. The optimum conditions for modulus of elasticity and porosity contents of final composite are projected and the infiltration characteristics under optimum conditions are determined and verified.

2. Experimental procedure

2.1. Taguchi design of experiments

Infiltration tests for Al alloys/SiC/SiO_2 composites were conducted under the conditions shown in Table 1. The effect of various parameters on the physical and mechanical properties of composite were investigated using the Taguchi method for design of experiments [15]. Infiltration temperature, time, particle size, porosity, SiO_2 phase type and content in the bilayer preform were used as parameters of infiltration process. These parameters were chosen because, according to a previous works, they have a substantial effect on the infiltration characteristics of SiC preforms by Al alloys [16]. Graded preforms were prepared by stacking of two SiC cylindrical preform layers of different particle size and porosity. In rows 3 and 4 of Table 1, the first number on the left corresponds to the value of the top preform layer. Accordingly, each of these parameters was tested at two or three levels. In addition, those parameter interactions that are perceived to have a significant effect on the properties were also included.

Table 1
Parameters and levels tested in the experiment.

Parameters	Level 1	Level 2	Level 3
Temperature	1100 °C	1200 °C	1300 °C
Time	90 min	120 min	150 min
SiC particle size	10 μm/129 μm	129 μm/10 μm	Mixture/mixture
Preform porosity	40/60	60/40	50/50
Crystallinity of SiO_2	Crystal (Type I)	Amorphous	Crystal (Type II)
Alloy (Mg concentration)	Alloy 1 (low Mg)	Alloy 2 (high Mg)	
SiO_2 content	10 wt.%	15 wt.%	20 wt.%
Interaction 1		Time × temperature ($t \times T$)	
Interaction 2		Porosity × time ($P \times t$)	
Constants	• N_2 flow rate: 30 cm^3/min • Amount of alloy: 20 g	• Geometry and size of cylindrical preform (for each layer: diameter:10 mm, height 5 mm)	

The parameter interactions considered in this experiment are the interaction between time and temperature ($t \times T$) and the interaction between porosity and time ($P \times t$). Table 2 shows the parameters, levels and interactions that are investigated in this work arranged in a standard Taguchi L27 orthogonal array.

2.2. Materials and procedure

The chemical composition of the two alloys used in the experiment is presented in Table 3. The structure and phase of silica derived from rice husk are completely dependent of calcining temperature, atmosphere and chemical treatment on rice husk before and after burning. Based on previous investigations [17,18], three procedures were chosen for extraction of silica from rice husk. The extraction procedure and physical properties of obtained silica corresponding to each method are summarized in Table 4.

Infiltration trials were performed in a horizontal tube furnace with a 6.5 cm diameter alumina tube closed at both ends with end-cap fittings to control the process atmosphere. Both fittings were sealed with O-rings and the system worked at a very slight over-pressure to that of the ambient. A K-type thermocouple was inserted in one end of the tube to record the sample temperature. The mold-preform-metal assembly was placed in the center of the alumina tube and heated in ultra-high purity nitrogen 30 °C/min up to test temperature. During heat up, any residual binder was volatilized and removed from the system in the flowing nitrogen. The chamber was held in an isothermal state for various test times in N_2 atmosphere. Finally the system was cooled still in nitrogen down to ~550 °C, after which the cooling to room temperature was carried out in the absence of nitrogen. After removing excess aluminum alloy from cylindrical composites, specimens were prepared for density measurement and mechanical property testing. Specimens for microstructure analysis were mounted and polished using standard metallurgical procedures, and the analysis was done using scanning electron microscopy (SEM), energy dispersive X-rays (EDXs), and X-ray diffraction (XRD). The modulus of elasticity of the composites was determined using an ultrasonic technique according to ASTM: E494-95. The thermal behavior of some selected samples were studied by differential thermal analysis (DTA). DTA curves were recorded on powdered samples in nitrogen at the heating rate of 20 °C/min, using an SDT Q600 thermo-analyzer with Al_2O_3 as reference material.

In order to investigate the wetting behavior of aluminum alloys on SiO_2 and SiO_2/SiC substrates, aluminum specimens, ~10 × 10 × 10 mm in size were cut from as-cast aluminum alloys. After cutting the specimens, they were ground on emery paper and ultrasonically washed in ethylic alcohol. Based on the test parameters, the prepared blocks were placed on the top of loose beds of SiC powders and of mixtures of SiC/SiO_2 (from rice husk) powders. The prepared specimen was placed into a horizontal tube furnace, which was then flushed with nitrogen (99.999% Ultra

Table 2
Standard L27 Taguchi table designed for pressureless infiltration process.

No.	A	B	C	D	E	F	G	H	I	Explanation
1	1	1	1	1	1	1	1	1	1	CD
2	1	1	1	1	2	2	2	2	2	CD
3	1	1	1	1	3	3	1•	3	3	CD
4	1	2	2	2	1	1	1	2	2	PI
5	1	2	2	2	2	2	2	3	3	PI
6[a]	1	2	2	2	3	3	1•	1	1	CD
7	1	3	3	3	1	1	1	3	3	CD
8	1	3	3	3	2	2	2	1	1	CD
9	1	3	3	3	3	3	1•	2	2	CD
10	2	1	2	3	1	2	1•	1	2	PI
11	2	1	2	3	2	3	1	2	3	CD
12	2	1	2	3	3	1	2	3	1	NI
13	2	2	3	1	1	2	1•	2	3	CD
14[a]	2	2	3	1	2	3	1	3	1	CD
15	2	2	3	1	3	1	2	1	2	NI
16[a]	2	3	1	2	1	2	1•	3	1	CD
17[a]	2	3	1	2	2	3	1	1	2	CD
18	2	3	1	2	3	1	2	2	3	NI
19[a]	3	1	3	2	1	3	2	1	3	CD
20	3	1	3	2	2	1	1•	2	1	NI
21	3	1	3	2	3	2	1	3	2	NI
22[a]	3	2	1	3	1	3	2	2	1	CD
23	3	2	1	3	2	1	1•	3	2	NI
24	3	2	1	3	3	2	1	1	3	NI
25	3	3	2	1	1	3	2	3	2	CD
26	3	3	2	1	2	1	1•	1	3	NI
27	3	3	2	1	3	2	1	2	1	CD

A: Temperature; B: Time; C: Temp. × time; D: Particle Size; E: Porosity; NI: Not Infiltrated; G: Alloy; H: Porosity × time; I: SiO_2 content; F: Phase of SiO_2; PI: Partially Infiltrated; CI: Fully Infiltrated.

[a] Specimens which degraded after infiltration process, within one or two weeks.