



# Processing of wood-derived copper–silicon carbide composites via electrodeposition

K.E. Pappacena, M.T. Johnson, S. Xie, K.T. Faber\*

Department of Materials Science and Engineering, Northwestern University, 2220 N. Campus Drive, Evanston, IL 60208, USA

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## ABSTRACT

An electrodeposition technique has been adapted to produce copper–silicon carbide composites with honeycomb-like microstructures. The detrimental Cu–SiC reaction was avoided by using this room temperature processing technique. The wood-derived silicon carbide phase allows for tailorable microstructures due to the variety of available wood precursors. Plating efficiency for each wood type was determined using image analysis. This processing method results in the successful filling of pores with aspect ratios of up to 100.

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## 1. Introduction

Electronic packaging, heat sink, and heat spreader materials are, in large part, used to remove heat from integrated circuits to prevent overheating and subsequent failure of the devices [1–3]. These materials need to have a combination of properties which include high thermal conductivity to dissipate heat quickly, low thermal expansion mismatch with the desired substrates, and low density to reduce weight. A recent trend has been to use composites for these applications, taking advantage of the high thermal conductivities of metals, such as copper, with the lower weights and thermal expansion coefficients of ceramics, such as silicon carbide.

In this paper we discuss a novel fabrication method for copper–silicon carbide composites for thermal management applications using wood-derived silicon carbide (biomorphic silicon carbide, or bioSiC). BioSiC takes advantage of the preexisting porous microstructure of wood as well as the variety of pore distributions among wood species to result in a processing method requiring lower temperatures and energy than traditional silicon carbide sintering [4–7]. BioSiC is characterized by elongated pores, parallel to the growth direction of the tree, which give it an anisotropic microstructure. The transverse direction is perpendicular to these long channels. The porosity in wood-derived materials follows a bimodal distribution. Larger pores result from vessels, which are used for transporting nutrients throughout the tree. Smaller pores result from fibers and rays, and are used for strength and nutrient storage [8]. The main challenge in producing copper–biomorphic silicon carbide (Cu–bioSiC) composites lies in the introduction of the me-

tal phase to the preexisting ceramic scaffold. Melt infiltration of the metal phase into the pores has been used previously to produce Al alloy–bioSiC composites [9–12]. Both copper and an aluminum alloy have been infiltrated into porous bioSiC by Lin et al. [13]. However, when molten copper comes into contact with silicon carbide, SiC is dissolved into Cu and excess carbon precipitates out until the Si concentration approaches its solubility limit, about 14.5 at.% at 1200 °C [14]. This reaction is detrimental to the thermal properties, as the addition of as little as 1.5% silicon to copper decreases the thermal conductivity by about 85% relative to pure Cu [15]. Thus, the Cu–SiC reaction is important to avoid when fabricating Cu–SiC composites.

A novel method to generate copper–silicon carbide (Cu–SiC) composites while avoiding the Cu–SiC reaction utilizes room temperature electrochemical deposition, thus eliminating the interaction of molten copper with silicon carbide. Yih et al. fabricated a Cu–SiC composite in which SiC whiskers were electrolessly plated and electroplated with copper prior to hot pressing at 950 °C, which is below the melting point of Cu, thus preventing the dissociation reaction [16]. Brendel et al. also used electroplating to coat SiC with Cu prior to hot isostatic pressing to 650 °C to make composite materials for fusion reactors [17]. Copper electrodeposition has also been performed in pores in anodized aluminum in order to make copper nanowire devices [18]. Olevsky et al. used sequential electrophoretic deposition and electroplating to first make a coating of porous alumina and subsequently impregnate copper into the pores, resulting in a composite resembling alumina particles in a copper matrix [19]. They also coated porous silicon carbide with a layer of copper using electrodeposition which resulted in enhanced thermal properties [19].

\* Corresponding author.

E-mail address: [k-faber@northwestern.edu](mailto:k-faber@northwestern.edu) (K.T. Faber).

Electrolyte solutions used for copper deposition often contain copper sulfate and sulfuric acid. In electronic applications, where it is common to electroplate into pores with high aspect ratios of up to 20, polyethylene glycol (PEG) and a source for chlorine ions ( $\text{Cl}^-$ ) are added [20–27]. These additives serve both to control the deposition rate, which leads to an even and smooth electroplated surface, and lower the surface tension of the electrolyte, which leads to increased solution penetration into the pores [20–27]. Solutions without additives result in void formation in the pores due to overfilling, leading to premature choke-off [27].

The examples of Cu–SiC composite fabrication using electrodeposition suggest that it is a feasible processing method to eliminate the detrimental Cu–SiC reaction. Using a basic electrolyte as described above, it is expected that electroplating can be applied to bioSiC samples and the challenge of deposition within the elongated pores, with aspect ratios of about 10–100 for samples with a pore length of 3 mm, can be overcome. We describe here the efficacy of electroplating as a fabrication method for Cu–bioSiC composites and investigate the Cu–SiC interface. The resulting Cu–bioSiC composites have potential for improved thermal properties, which are demonstrated in an additional paper [28], and thus may prove useful in thermal management applications.

## 2. Experimental methods

### 2.1. Processing of copper–bioSiC composites

Four hard wood precursors were used to fabricate silicon carbide scaffolds for copper–silicon carbide composites: beech (*Fagus sylvatica*), poplar (*Liriodendron tulipifera*), red oak (*Quercus rubra*) and sapele (*Entandrophragma cylindricum*) [29]. Carbon was made by pyrolyzing samples of each wood type to 1000 °C in argon. A scanning electron micrograph of a cross-section of sapele-based carbon is shown in Fig. 1a. Following pyrolyzation, the porous channels of the carbon samples were infiltrated with molten silicon in a boron nitride-coated alumina crucible at 1500 °C under vacuum [4–7]. After infiltration, excess silicon remaining in the pores was removed by placing the samples in a continuously stirring nitric/hydrofluoric acid etch for 1–2 weeks, as described by Kaul et al. [30]. Fig. 1b shows a scanning electron micrograph of a cross-section of sapele-based bioSiC. It is clear from Fig. 1a and b that the wood microstructure is retained throughout the pyrolyzation and infiltration steps. The microstructures of the initial wood, the pyrolyzed carbon, and the resulting bioSiC are anisotropic, and are characterized by elongated pores.

Preliminary experiments to make composites were performed using melt infiltration of copper into the pores of the bioSiC. Copper was melted onto bioSiC samples at 1200 °C for 30 min in argon. These samples were used to study both the ability of the Cu to wet

and infiltrate the pores of the bioSiC and the reaction behavior of molten copper and bioSiC.

In a second series of experiments, copper was electrodeposited on bioSiC samples using a plating electrolyte described by Kelly and West [21]. The components of this bath were 0.24 M  $\text{CuSO}_4 \cdot 5\text{H}_2\text{O}$ , 1.8 M  $\text{H}_2\text{SO}_4$ , 300 mg/mL 3400 MW PEG, 0.05 mg/mL chloride ions in the form of HCl, and the balance was deionized water. Prior to adding samples to the solution, they were vacuum infiltrated with the electrolyte, encouraging the solution to coat the inner pore surfaces of the material. BioSiC samples, which had electrical conductivities of about 0.75 Ohm, [31] were negatively charged by connecting them to an external power supply using alligator clips. Due to the variation in resistance with copper deposition and depletion of the electrolyte, the power supply was operated at a constant current of 25 mA to maintain a constant plating rate.

The samples remained in the continuously stirring electrolyte bath for 5 days, with one additional vacuum infiltration between the second and third day of plating. Electroplated samples were 12.7 mm in diameter and 3 mm thick. They were fabricated in either the axial or the transverse orientation as illustrated in Fig. 2. Several Cu–bioSiC samples were heated to 1000 °C in argon for 30 min to test for any Cu–SiC reaction.

In red oak-based Cu–bioSiC composites, the larger pores (>100  $\mu\text{m}$ ) were choked off and left incompletely filled when this protocol was used. The processing conditions were altered by first lowering the current to 10 mA to slow the deposition rate and prohibit the choking off of pores. Additionally, the time the sample remained in the bath was increased to accommodate the slower plating rate. The red oak samples remained in the bath for 7 days, until a continuous copper layer was present on the surface. This outer layer was ground off using 180–240 grit SiC paper to expose the pores that had been choked off. The sample was then vacuum infiltrated with fresh electrolyte and placed in a fresh solution for another 5 days to allow these pores to fill more completely.

### 2.2. Characterization methods

Scanning electron microscopy (Hitachi S-3400N-II SEM, Hitachi High Technologies America Inc., Pleasanton, CA) was used to investigate the microstructure of the Cu–bioSiC composites. Elemental distributions were determined using energy-dispersive X-ray spectroscopy (EDS). The backscattered electron detector (BSE) was also utilized to readily identify the copper, silicon carbide, and pore phases in composite samples.

Grazing incidence parallel beam X-ray diffraction measurements were performed using a Rigaku ATX-G diffractometer (Rigaku Americas, The Woodlands, Texas). The ATX-G was operated at 50 kV and 240 mA, using  $\text{Cu K}\alpha$  radiation (1.54 Å). A  $2\theta$  scan was performed at  $4^\circ \text{min}^{-1}$  with a step size of  $0.05^\circ$ , an incident beam width of 0.1 mm, and the sample tilt angle fixed at  $0.6^\circ$ .

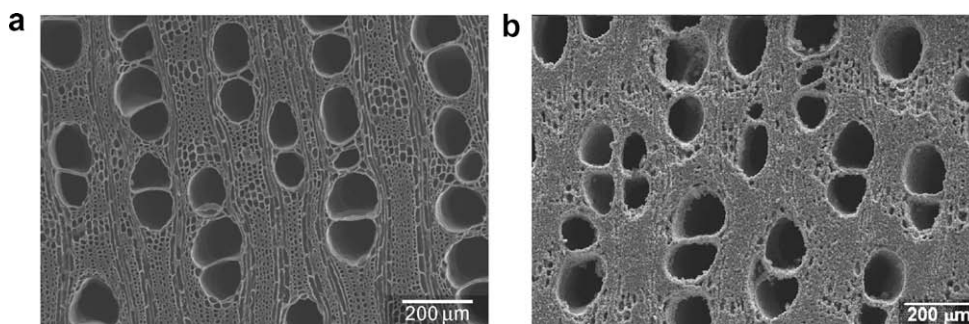


Fig. 1. Scanning electron micrographs of (a) sapele-based carbon and (b) sapele-based silicon carbide.

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