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# Diffusion bonding of SiC fiber-bonded ceramics using Ti/Mo and Ti/Cu interlayers

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**Abstract**

A SiC fiber-bonded ceramic (SA-Tyrannohex™) was diffusion bonded using Ti/Mo and Ti/Cu interlayers. The influence of metallic interlayers and SiC fiber orientation in the ceramic substrate with respect to the interlayers on joint microstructure, elemental composition, and microhardness in diffusion bonds was investigated using Optical Microscopy (OM), Field Emission Scanning Electron Microscopy (FE-SEM), Energy Dispersive Spectroscopy (EDS), and Knoop microhardness test. Compared to the Ti/Mo bilayers, the Ti/Cu bilayers yielded higher quality joints. The reaction products distributed more homogeneously across the joint thickness in Ti/Cu bonds than in Ti/Mo bonds. The reaction layers adjacent to the SiC substrate in both parallel and perpendicular SA-THX/Mo/Ti/SA-THX joints were twice as hard as the joint center where the Mo interlayer had remained untransformed during diffusion bonding. In SA-THX/Cu/Ti joints, hardness distribution was uniform across the joint thickness consistent with a more homogeneous reaction phase distribution across the joint.


*Keywords:* B. Electron microscopy; Diffusion bonding; Silicon carbide; Knoop hardness; Reaction layers

## 1. Introduction

Silicon carbide (SiC) based materials have excellent high-temperature mechanical properties, oxidation and heat resistance, and thermo-chemical stability. These materials are leading candidates for various thermostructural applications at high temperatures in harsh environments in aerospace and energy sectors [1–3]. A variety of advanced SiC-based ceramics such as CVD SiC, sintered SiC, hot-pressed SiC, and SiC fiber-bonded ceramic (SA-Tyrannohex™) as well as ceramic composites such as SiC/SiC and C/SiC have been developed for such applications. Implementing these advanced ceramics and composites in real components demands robust ceramic joining and integration technologies. The most commonly used ceramic joining methods include reaction bonding [4] and brazing [5,6]. Diffusion bonding techniques have been successfully utilized to bond a wide variety of SiC ceramics. Sintered, hot-pressed and CVD silicon carbide ceramics have been diffusion bonded using refractory metal interlayers of titanium [7–10], molybdenum [11,12], tantalum [13], tungsten [14], niobium [13], zirconium [10], nickel [15] and Inconel 600 [16]. The diffusional transformation of a metal interlayer at high temperatures and under high mechanical pressures into carbides, silicides, or complex ternary and higher order compounds produces strong joints. Carbides and silicide compounds of refractory metals are also thermodynamically more stable than SiC. As a result, diffusive conversion of metal insert into carbides and silicides provides a pathway for strong bond formation.

Previously, some of the present authors [17–19] investigated the diffusion bonding of SiC/Ti/SiC joints and identified the phases that form in the bonded area using scanning electron microscopy (SEM), X-ray diffraction (XRD) analysis, and energy dispersive spectroscopy (EDS). Recently, the authors conducted a detailed transmission electron microscopy (TEM) study for the structural evaluation of the phases formed during

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diffusion bonding with Ti as the interlayer [20,21]. All prior studies investigated diffusion bonding of sintered, hot pressed, and CVD SiC ceramics, and apart from the present authors' recent research [22] on diffusion bonded SA-Tyrannohex™ (also referred to as SA-THX) using Ti interlayers, no published reports currently exist on diffusion bonding of SiC fiber-bonded ceramic, SA-THX. The liquid-phase joining of SA-THX via brazing was also only recently demonstrated [23–25].

SA-THX is a dense and tough ceramic consisting of a highly ordered, closed-packed structure of fine hexagonal columnar fibers composed of crystalline β-SiC with a thin (10–20 nm) interfacial layer of turbostatic carbon between the fibers [26,27]. SA-THX is a product of Ube Industries, Japan, and is obtained by converting an amorphous Si–Al–C–O fiber into Tyranno-SA fiber with near-stoichiometric composition. The final SiC fiber-bonded ceramic, SA-THX, is made by hot-pressing of fiber weave lay-ups into various configurations. The resulting material exhibits good thermomechanical performance, high thermal conductivity (33 W/m K at 1600 °C), high strength up to 1600 °C, high flexural strength (~300 MPa), high fracture toughness (1200 J/m$^2$ at room temperature), and good machinability and low cost.

In this study, SA-THX was diffusion bonded to itself using either Ti/Mo or Ti/Cu bilayers to examine the effect of the interlayers and SiC fiber orientation (perpendicular and parallel relative to the joining plane) on the microstructure, composition and hardness of joints using Optical Microscopy (OM), Field Emission Scanning Electron Microscopy (FE-SEM), Energy Dispersive Spectroscopy (EDS), and the Knoop microhardness test.

## 2. Experimental

SA-Tyrannohex™ (SA-THX) SiC fiber-bonded ceramic was obtained from Ube Industries (Ube, Japan). The SA-THX is made from SA-Tyranno fiber bundles woven into an eight-harness satin weave that results in fibers being oriented in parallel (||) and perpendicular (⊥) directions. Joints were made using like SA-THX composite pairs (||-to-||, or ⊥-to-⊥). Ti foil (10 μm), Mo foil (12.7 μm), and Cu foil (5 μm) were obtained from Goodfellow Corporation (Glen Burnie, MD, USA). Before joining, all materials were ultrasonically cleaned in acetone for 10 min. Joints were diffusion bonded at 1200 °C with a pressure of 30 MPa. Joint processing was conducted in vacuum for 4 h at the peak temperature under load, followed by cooling at a rate of 2 °C per min. Joints with Ti/Cu bilayers had three Cu interlayers (total thickness: 15 μm) with 10 μm thick Ti foil on two sides. Similarly, for Ti/Mo bilayers, a 12.7 μm Mo foil was sandwiched by 10 μm thick Ti foil on two sides. The nomenclature used to identify the joints for subsequent discussion is as follows. A SA-THX joint with Ti and Mo interlayers having ⊥fibers coincident on the foil is denoted as follows: ⊥SA-THX/Ti/Mo/Ti/⊥SA-THX. Similarly, a SA-THX joint with || fibers coincident on the foil is denoted as: || SA-THX/Ti/Mo/Ti/|| SA-THX.

Optical Microscopy (OM) was done on an Olympus BX51, and SEM observations and elemental analyses were performed with a Hitachi S-4700-I Field Emission Scanning Electron Microscope (FE-SEM) coupled with an Energy Dispersive Spectrometer (EDS). Joint hardness was characterized using Knoop indenter on a Struers Duramin-A300 machine under a load of 200 g and loading time of 10 s.

## 3. Results and discussion

### 3.1. Joint microstructure and composition

#### 3.1.1. SA-THX/Ti/Mo/Ti SA-THX joints

In designing the SA-THX joints using Ti/Mo bilayers, dual objectives were sought to be accomplished. One objective was to take advantage of a low coefficient of thermal expansion (CTE) of Mo interlayer (CTE: $5.1 \times 10^{-6}\,K^{-1}$) to reduce residual stresses in diffusion bonded joints. A small CTE mismatch mitigates the residual stress buildup from joining. Experience has shown that judiciously chosen interlayers positioned in the joint prior to bonding can reduce the strain energy and cracking propensity in ceramic joints. A second objective was to explore the feasibility of lowering the bonding temperature relative to direct bonding using only Mo foils. Past research [11,14] has shown that SiC/Mo joints diffusion bonded under proper conditions have excellent bond quality; in fact, the quality of the SiC/Mo/SiC bond was significantly better than the diffusion bonds produced with Ti. Cockeram [14] observed that high temperatures (1500 °C) and long contact times (10 h) produced sound SiC/Mo joints under 3.4 to 18 MPa pressures with 12.7 μm thick Mo foils. Similar joints processed at 1200 °C led to poor bond quality. In Ref. [14], it was also noted that use of thicker molybdenum foil (25.4 μm) resulted in a significantly higher density of cracks in the bond region in a manner similar to the diffusion bonding of SiC using Ti where use of thin rather than thick foils improved the quality of the interfacial bond [14]. Thus although Mo (CTE: $5.1 \times 10^{-6}\,K^{-1}$) and SiC are better CTE matched than are Ti (CTE: $8.6 \times 10^{-6}\,K^{-1}$) and SiC, thin Ti (10 μm) and Mo (12.7 μm) metallic foils were both used together in the present work with the goal to achieve sound diffusion bonds at relatively low (1200 °C) operating temperatures.

Titanium and molybdenum form an isomorphous binary alloy system and Ti and Mo are completely miscible above 882 °C. Below 882 °C, the solubility of Mo in Ti is restricted (e.g., the maximum solubility of Mo in α-Ti is less than 0.5% at 600 °C [11]). It was envisioned that formation of a Ti(Mo) solid solution in conjunction with the carbides and silicides from the reactions between Ti and SiC and between Mo and SiC shall reduce the residual stresses in SiC joints while still forming strong bonds and lowering the energy required for diffusion bonding as a result of reduced process temperatures (from 1500 °C to 1200 °C).

A 12.7 μm Mo foil was sandwiched by 10 μm Ti foils on two opposite sides in pairs of parallel and perpendicular substrates. Optical images of parallel (|| SA-THX/Ti/Mo/Ti/|| SA-THX) and perpendicular (⊥ SA-THX/Ti/Mo/Ti/⊥ SA-THX) joints are shown in Fig. 1a and b, respectively. The ends