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INFLUENCE OF MATERIAL COATING ON THE HEAT TRANSFER IN A LAYERED Cu-SiC-Cu SYSTEMS

This paper describes the process of obtaining Cu-SiC-Cu systems by way of spark plasma sintering. A monocrystalline form of silicon carbide (6H-SiC type) was applied in the experiment. Additionally, silicon carbide samples were covered with a layer of tungsten and molybdenum using chemical vapour deposition (CVD) technique. Microstructural examinations and thermal properties measurements were performed. A special attention was put to the metal-ceramic interface. During annealing at a high temperature, copper reacts with silicon carbide. To prevent the decomposition of silicon carbide two types of coating (tungsten and molybdenum) were applied. The effect of covering SiC with the aforementioned elements on the composite's thermal conductivity was analyzed. Results were compared with the numerical modelling of heat transfer in Cu-SiC-Cu systems. Certain possible reasons behind differences in measurements and modelling results were discussed.

Keywords: copper matrix composites, silicon carbide, interface, thermal conductivity, modelling

1. Introduction

Multilayer materials combining metallic and ceramic components can be widely used in electronic, power and optical applications [1]. The removal of heat generated in electronic power devices (e.g., diodes and lasers) during exploitation is crucial for their operational effectiveness. The components of such substrates should be characterized by a high thermal conductivity and the thermal expansion coefficient matched with the properties of semiconductors. The mismatch of mechanical and thermal properties for ceramic and metallic materials can cause a high level of residual stresses to occur [2-4]. The combination of copper and silicon carbide seems to be a good solution with great scientific and technological potential. In the case of Cu-SiC systems it is very important to avoid some disadvantageous phenomena occurring at Cu-SiC boundary that can affect thermal properties. It should be noticed that copper decomposes SiC to Si and C at an elevated temperature [5].

The subject literature lists a couple of analytical methods leading to strict solutions of issues related to the transient heat transfer in layered composite materials, including: (i) the orthogonal or quasi-orthogonal expansions method, a particularly effective method in case of transient heat transfer in multilayer systems with a finite thickness without any internal heat sources [6-8], (ii) the Laplace transform method – a convenient method for solving the issues of transient heat transfer in layered systems comprising layers with an infinite thickness, (iii) the Green's function method – applied in initial-value boundary problems of the transient heat transfer in layered composites with internal heat sources [9]. In addition, there are a number of approximation methods to solve the issues of transient heat transfer, e.g. integral transform method over a finite area or variation methods.

This article describes a numerical model of heat transfer through a layered composite material in order to define effective thermal properties of the said composite depending on the construction of the interface at the metal-ceramic boundary. Compared to analytical methods, numerical modelling allows for the analysis of complex-shaped materials, composites with different component materials, different types of such materials and with different configurations, while analytical methods are mainly applied to specific shapes (e.g. spherical inclusions in a matrix) as well as to a limited number of component materials. In this paper technological trials to obtain Cu-SiC-Cu layered systems were conducted. The investigation of the manufactured materials was aimed at: (i) determining structural changes occurring at the metal-ceramic boundary, (ii) determining the effect of SiC surface modification on the system's thermal conductivity, (iii) establishing a numerical model of heat transfer in a multilayer system.

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2. Experimental procedure

Copper powder (with the average grain size of 40 µm, NewMet Koch) with a commercial purity of more than 99.99% was used as the starting material. A 6H-SiC crystal from which plates as thin as 1.0 mm were carved, had been obtained from the vapour phase by means of the Physical Vapour Transport (PVT) method and was crystallised at the temperature of 2150°C. Molybdenum and tungsten layers with the thickness of 0.8 µm were deposed on both sides of the 6H-SiC monocrystal with the use of the Chemical Vapor Deposition (CVD) method. Prior to vacuum spraying, 6H-SiC plates were prepared, namely washed in concentrated non-organic acid at a hot temperature, and then their surfaces were deoxidized in HF solution. The process of deposing the layers of W and Mo was carried out with the use of a device by ULVAC at vacuum of 1-2×10^{-2} mbar with the use of 300 W generator for 100 min. Cu-SiC-Cu layered composites were prepared as provided on the schematic below (Fig. 3) and subjected to the SPS process. In a graphite matrix SiC plate was placed on copper powder, while its top surface was covered with copper, too. The spark plasma sintering process (SPS) was conducted in a vacuum atmosphere (5×10^{-5} mbar) using the following parameters: sintering temperature = 950°C, heating rate = 100°C/min, holding time = 10 min, and pressure = 50 MPa. Finally the rectangular samples with dimensions 8.0×8.0×3.0 mm were obtained. Following sintering, the thickness of particular component layers in the Cu-SiC-Cu system was 1 mm.

Next, the microstructure and thermal diffusivity of the manufactured Cu-SiC-Cu systems were examined. The microstructure of the Cu-SiC-Cu layered systems were examined using a scanning electron microscopy AURIGA CrossBeam Workstation (Zeiss). Thermal conductivity was measured at the temperature range 50-300°C using the Laser Flash Analyser LF457/Netzsch in argon atmosphere. The measured data was analyzed taking both heat loss and finite-pulse effects into consideration. Standard constituents of the analysis software are the improved Cape-Lehmann model, the possibility of considering for radiative heat transfer, and non-linear regression routines for the characterization of two and three-layer systems.

For the calculation of the thermal conductivity of multilayer composite a FEM (finite element model) using a commercial program FEAP ver. 7.5 was developed. 2D, 9-nodes thermal elements which solve the linear Fourier heat conduction equation in a two-dimensional domain were used Eq. (1):

$$q = -k_{eff}\,\Delta T \tag{1}$$

where: q is the thermal flux, k_{eff} is the effective thermal conductivity and $\Delta T = T_{hot} - T_{cold}$.

The temperature on the two opposite faces of the specimen was constant but one face was set as hot (T_{hot} = 100), the opposite face was kept cold (T_{cold} = 0), whereas the remaining ones were assumed to be adiabatic. Following [3], the effective thermal conductivity was described by Eq. (2):

$$k_{eff} = -\frac{Q_1}{A}\frac{L}{(T^1_{hot} - T^1_{cold})} \tag{2}$$

where: L is the length of the square side, A – the cross-section area of the square ($A = L^2$), $T_{hot} - T_{cold}$ is the temperature difference across the unit cell, Q_1 – the overall heat transfer into the unit cell.

3. Results and discussion

Observations by means of scanning electron microscopy were made to assess the quality of the obtained Cu/SiC bonding at the boundary of both phases. Figure 1a shows the typical microstructure of Cu-SiC-Cu layer system with uncoated SiC after the SPS process. In order to ensure a high thermal conductivity of the entire system, the most crucial thing is to obtain a strong metal-ceramic bonding, preferably – adhesive one. The changes in the chemical composition of this area may decrease the material's capacity to transport heat (the dissolution of silicon in copper in particular). Observations of the Cu/SiC interface with uncoated SiC (Fig. 1a) with scanning electron microscopy revealed that as a result of a chemical reaction, silicon carbide is decomposed.

The layer of copper that adheres to SiC becomes enriched with silicon that dissolves in it. The range of visible changes reaches 2 µm. On the surface, spherical precipitations can be seen – probably Cu_3Si. The interface reaction ($Cu + SiC = Cu_3Si + C$) can easily take place, which is detrimental to the thermo-physical properties due to the solid solution of Si and formation of residual graphite [10]. Such changes largely decrease the thermal conductivity of copper, and – in what follows – of the whole

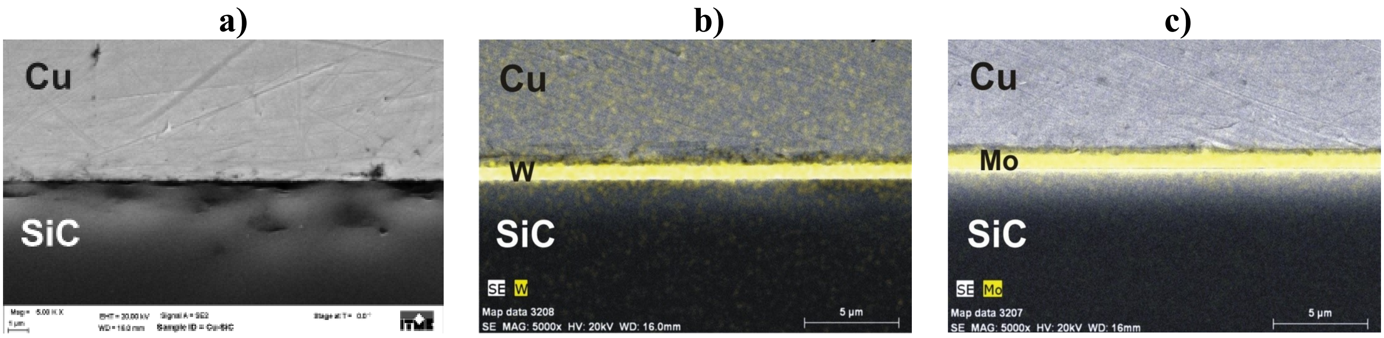

Fig. 1. SEM micrographs of Cu-SiC boundary: a) Cu-SiC b) Cu-SiC(W) and c) Cu-SiC(Mo)

system. In addition, discontinuities and local porosity at the metal-ceramic boundary can be observed, which is detrimental to mutual adhesion at the boundary of both phases and causes the physical and mechanical properties to decrease. Fig. 1b and 1c presents SEM micrographs of Cu-SiC-Cu systems where silicon carbide had been covered with molybdenum or tungsten prior to the manufacturing process.

The observation of Cu/SiC interface has shown that the thickness of the applied layers of molybdenum and tungsten is not uniform on the cross-section of the whole bonding, and ranges from 0.68-0.77 μm for Mo and 0.74-0.88 μm for W. What is more, one can observe that the obtained layer is continuous, has no visible defects in the near-surface layer of SiC, which proves that the use of coating prevents silicon carbide from decomposition. Tungsten and molybdenum with copper do not show mutual solubility neither as liquids, nor as solids, thus they provide an ideal covering ensuring a hermetic protective layer of SiC.

The results of thermal conductivity measurements for three systems of Cu-SiC-Cu composite in the function of temperature are shown in Figure 2.

The obtained results of thermal conductivity of layered composites has shown that covering a SiC monocrystal with a layer of molybdenum or tungsten results in higher values of thermal conductivity compared to the uncoated SiC system. As expected, better results were obtained in case of using tungsten rather than molybdenum.

Based on the structural analysis of the Cu/SiC interface a numerical model of heat transfer in Cu-SiC-Cu layered composites was established. The first step in modelling was to create the model of composite without an additional layer (W or Mo).

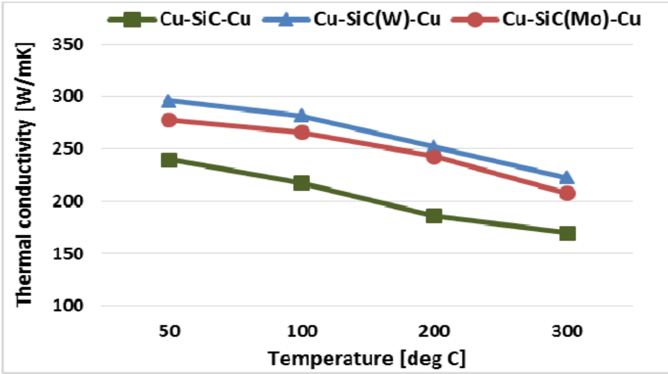

Fig. 2. Thermal conductivity of Cu-SiC-Cu system in temperature range 50-300°C

The FE mesh prepared for this case is presented on Figure 3. In order to take into account the changes of thermal conductivity associated with the diffusion of Si atoms into copper we assumed, in numerical model a thin layer (about 0.8 mm) with thermal conductivity of mixed value of Si an Cu (green colour). Also, we observe some pores on the interface of Cu and SiC – in the numerical model a small porosity on the interface is added to take this into account (5% of interface we assumed to be pores, yellow colour on Fig. 3a). Next, we use FE model to numerically calculate the effective thermal conductivity of 5 layered systems Cu-SiC(W)-Cu and Cu-SiC(Mo)-Cu. The schematic FE mesh is presented in Figure 3b.

As it can be seen in the Fig. 4, the numerically calculated thermal conductivity is higher for all systems than the one measured experimentally. The difference ranges reaches about 10%.

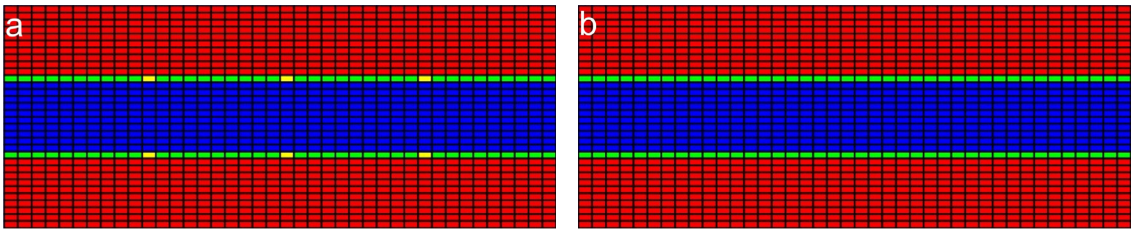

Fig. 3. The Finite Element Mesh used for numerical calculation. Red colour represent Cu, blue is SiC, green is interface layer and yellow represent pores (only on Fig. 3a)

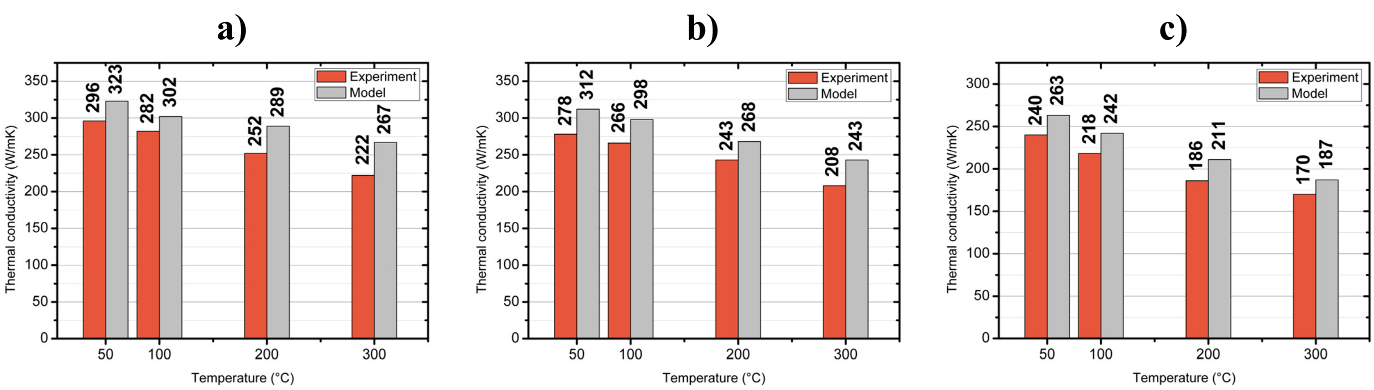

Fig. 4. Thermal conductivity for: Cu-SiC(W)-Cu system (a), Cu-SiC(Mo)-Cu system (b) and Cu-SiC-Cu system (c) in temperature range – experimental data vs. modelling calculation

This can be explained by approximations used for the sake of analysis. In numerical models, only for Cu-SiC-Cu system we assumed some discontinuities in the interface between Cu and SiC. For other composites, the ideal interface was assumed. In real systems some discontinuities always exist – not included here in the model (e.g. local porosity, homogenous dissolution of silicon in copper in the whole thickness of the defectated layer and a uniform cover of SiC with metal). This is what was expected, because of a higher thermal conductivity of tungsten than molybdenum. Regardless of the differences, in terms of quality the consistence with data provided by the subject literature was obtained. The importance as well as righteousness of using a protective coating on the surface of silicon carbide were confirmed.

4. Conclusions

It has been identified in case of Cu-SiC-Cu systems that there is a problem with interfacial reaction which has an effect on the material's properties. In order to improve the quality of boundary, the SiC was coated with a thin tungsten or molybdenum layers using CVD technique. The coating was used to prevent the decomposition of SiC. Microstructural observation of the metal-ceramic interface in Cu-SiC-Cu layered systems has shown that in case of pure SiC, the Cu/SiC bonding is weak, discontinued and porous. The use of coating on SiC - a thin layer of molybdenum or tungsten has allowed to prevent the decomposition of silicon carbide as well as to obtain a continuous, non-defectated metal-ceramic bonding. According to thermal conductivity results it has been shown that the use of tungsten or molybdenum coating on a SiC monocrystal results in a significant improvement of thermal conductivity of the analysed systems. Higher thermal conductivity results were obtained for tungsten. The results of the numerical analysis have confirmed the results of real systems measurements, and the reported differences are caused by approximations adopted in the analysis with regard to the Cu/SiC interface.

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