MECHANICAL CHARACTERIZATION AND DAMAGE EVOLUTION OF A CU/SIC – INTERPENETRATING METAL MATRIX COMPOSITE MANUFACTURED BY A SQUEEZE-CAST INFILTRATION METHOD

A.Reeb[1], A.Wanner[2], K. A. Weidenmann[3]

Institute for Applied Materials (IAM-WK), Karlsruhe Institute for Technoloy (KIT) , Kaiserstr. 12, 76131 Karlsruhe, Germany
Email[1]: andreas.reeb@kit.edu, Web Page: http://www.iam.kit.edu/wk
Email[2]: alexander.wanner@kit.edu, Web Page: http://www.iam.kit.edu/wk
Email[3]: kay.weidenmann@kit.edu, Web Page: http://www.iam.kit.edu/wk

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Abstract
The properties of interpenetrating metal-ceramic composites (IPC) can be specifically tailored for application requirements, such as high thermal conductivity combined with low thermal expansion, as well as high stiffness and high strength. Therefore, they represent a promising material class for thermally stressed components e.g. heat sinks. This contribution focuses on the characterization of the mechanical properties of a Cu/SiC-interpenetrating composite manufactured by a high temperature squeeze casting process (HTSC). The microstructure of the composite was analyzed by means of light microscopy and SEM analysis. A homogenous distribution of the ceramic phase in the copper matrix was found, and also a significant amount of porosity caused by the manufacturing process was observed. The mechanical properties of the composite were investigated by Ultrasonic-Phase-Spectroscopy (UPS) and quasi-static compression tests. For a better understanding of the deformation and damage behavior under compression, in situ SEM compression tests, were performed. While Young's modulus showed only slight increase, compression strength and yield strength were increased significantly compared to pure copper. The in situ damage analysis shows interface delamination in loading direction, before reaching the compression strength of the composite and subsequent brittle failure of the specimen, due to merging of interfacial cracks.

1. Introduction

Copper offers high corrosion resistance, easy processing and is used for cooling of electrical and electronic components mainly due to its high thermal conductivity [1]. Thus it is one of the most common materials used for heat conductors and heat sinks. However, the use of copper at elevated temperatures is often restricted by its high thermal expansion and by its limited mechanical properties [2]. For this reason, more adapted materials for dissipation of the resulting heat are required, as a consequence of advancing densification of electrical components. In copper matrix composites, the high thermal conductivity of copper can be combined with the low thermal expansion of a second phase, which additionally acts as a reinforcing phase and improves the mechanical properties. In addition the metallic phase can be responsible for retaining some plasticity in the composite. As reinforcing phase in MMCs silicon carbide (SiC) is commonly used due to its high hardness and strength [3]. It also offers a good oxidation resistance and high chemical stability [2] and in respect to the resulting thermal conductivity of the composite it offers a high thermal conductivity of 200-300 W/mK and a thermal expansion coefficient of 4.5×10^{-6} K^{-1}. Metals with considerably higher melting temperatures, such as copper, are commonly processed by gas pressure infiltration or powder

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metallurgical processes [4] [5]. Squeeze cast infiltration of ceramic preforms with open porosity [6] could be a cost-effective production route with short processing time, which provides the benefit of a resulting fine microstructure [7][8]. This leads to higher material strength compared to other casting methods [9][10]. Furthermore the contact time between SiC and molten Cu can be kept short to avoid interface rections, which negatively effect thermal and electrical conductivity and reduce mechanical properties due to dissolution of SiC [11]. Yet, on laboratory scale, only few investigations with pressure assisted casting methods for copper were performed [10][12] and no further mechanical analysis of Cu/SiC-composites with interpenetrating structure are available . This work aims to give a first characterization of the mechanical properties and an understanding of the damage behavior of this composite to allow identifiying starting points for material optimization. Therefore, this investigation focuses on mechanical properties in elastic and elastic-plastic loading and the resulting damage behavior of a Cu/SiC-composite with an interpenetrating network structure fabricated by high temperature squeeze cast infiltration (HTSC). The mechanical characterization is accompanied by quantitative and qualitative analysis of the microstructure by means of SEM and light microscopy analysis. The elastic properties are determined by ultrasonic phase spectroscopy (UPS) according to [13][14].The elastic-plastic properties are investigated by means of quasi-static compression tests and the damage behavior is analyzed in detail by using an in situ compression test method [15], which allows quasi-static loading of samples inside a SEM.

2. Materials and Experimental setup

2.1. Materials

Commercially pure copper was used as matrix material for manufacturing the MMC by squeeze casting. The chemical composition is specified in Table 1. The copper used was of OF-quality, which means high quality, non-deoxidized, oxygen-free copper suitable for electronic devices because of its high purity and conductivity. The reinforcing ceramic structure was SiC in form of open porous preforms manufactured by isostatical pressing of SiC-powder. The synthesis of the SiC-prefroms is described in detail in [16].

Table 1 Chemical Analysis of copper used for infiltration in wt.-ppm

	Pb	Bi	As	Sb	Sn	Zn	Mn	Cr	Co	Cd	Fe	Ni	Ag	S	Se	Te	O
Copper	<1	<0.5	1	1	<0.5	<1	<0.5	<1	<1	<1	≤6	<2	0.1	4	<0.5	<0.5	≤5

The synthesis of Cu-SiC composites was done at Materials Research Insitute Aalen (IMFAA, Aalen University) using a high-temperature squeeze castig device (HTSC, FCT Systeme GmbH). This allows a controlled direct pressure onto the copper melt via an upper punch made of heat resistant steel. The casting die and the lower punch are made of nickel based alloy (Haynes®230). Preform temperature was chosen to be 1190 °C and temperature of the copper melt was 1300 °C. A detailed description of this squeeze casting process is given in [16].

2.2 Experimental setup

Sample preparation

Several cubic samples were cut out from the fabricated material for UPS-measurements and compression tests. For compression test and ultrasonic analysis, 5 x 5 x 5 mm³ cubic samples were used. To achieve a good coupling of the ultrasonic wave transducers, the surfaces of the specimen were polished with 1000 - 4000 SiC abrasive paper. For the in situ SEM compression tests cubic samples of 2 x 2 x 2 mm were cut out from the composite. The observed surcface was grinded with SiC abrasive paper (1000 - 4000) and additionally polished with diamond suspension (9 µm- 3 µm). The surfaces clamped into the testing rig were manually grinded with 4000 SiC abrasive paper. This

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preparation was done with a Gatan Disc Grinder which allowed manual grinding and aimed for a good parallelism of the polished surfaces.

UPS measurements
Ultrasonic phase spectroscopy (UPS) was used to determine the longitudinal and the shear wave velocities along all three spatial directions. This technique is based on the measurement of the phase shift occurring as continuous, sinusoidal, elastic waves propagating through the sample. This phase shift is recorded as a function of frequency and enables the calculation of the elastic constants. A detailed description of this method is given in [14] and [17]. In this contribution, an electronic network analyzer (Advantest, model R3754A) and two identical broadband ultrasonic longitudinal wave transducers (Panametrics, model V122 with nominal central frequency 7.5 MHz) and transverse wave transducers (Panametrics, model 155, with nominal central frequency 5 MHz) were used for the measurements. The transducers were attached to the opposite sides of the samples using a water soluble couplant. The phase and the amplitude spectra were recorded in the frequency range from 10 kHz to 15 MHz for longitudinal wave measurements and in the range from 10 kHz to 8 MHz for transverewave measurements.

Compression tests
The compression tests were conducted on a Zwick universal testing machine with a maximum load capacity of 500 kN. Testing speed was 0.15 mm/min resulting in a nominal strain rate of $5 \cdot 10^{-3}\,s^{-1}$.Strain measurement was done by using a capacitive strain gauge. The experimental setup is depicted in Fig. 1a. The termination of compression test was set to a maximum deformation of 1.5 mm to avoid damage of the capacitive sensor.

In situ SEM compression tests
In situ compression tests were carried out using a miniature mechanical testing system built by Kammrath & Weiss GmbH with a maximum load capacity of 10 kN (Fig. 1b). The total strain was measured with an in-built LVDT device (Linear Variable Differential Transformer). All experiments were carried out with a constant crosshead velocity of 0.2 µm/s, resulting in a nominal strain rate of $10^{-3}\,s^{-1}$ allowing a live view of the sample during loading with a reasonable sampling rate in the SEM and strain rates comparable to the macroscopic compression tests. The test rig was placed inside the vacuum chamber of a SEM of type Zeiss EVO 50 and allowed observation of the damage evolution in detail by suspending the test at different loads.

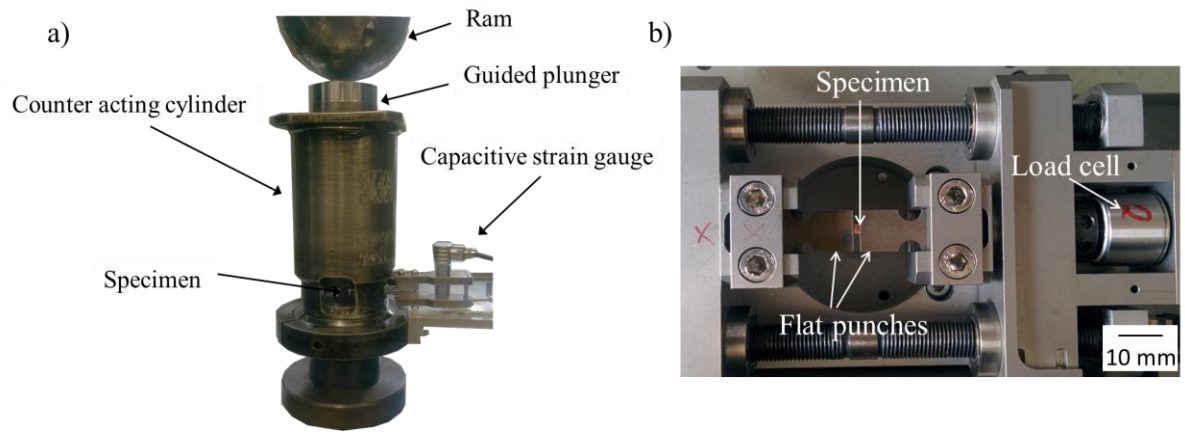

Figure 1. Experimental setup for a) compression tests and b) In situ SEM compression tests

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3.Results

The microstructural studies revealed a homogeneous distribution of the SiC phase in the sample (Fig. 2), but also a significant porosity was observed (Fig.2a). In particular, at the Cu/SiC-interface microporosities between copper phase and SiC phase can be recognized (marked in Fig. 2b). The density of the specimens was measured by using Archimedes priniciple and was determined to an average of 4.66 ± 0.2 g/cm³.The SiC-content was determined using digital image analysis (analysis pro V.5.1) and gives an approximated SiC-volume fraction of about 58.4 vol.- % . This leads to a overall porosity fraction of about 15.4 vol.-%, calculated with the measured mean density of the specimens.

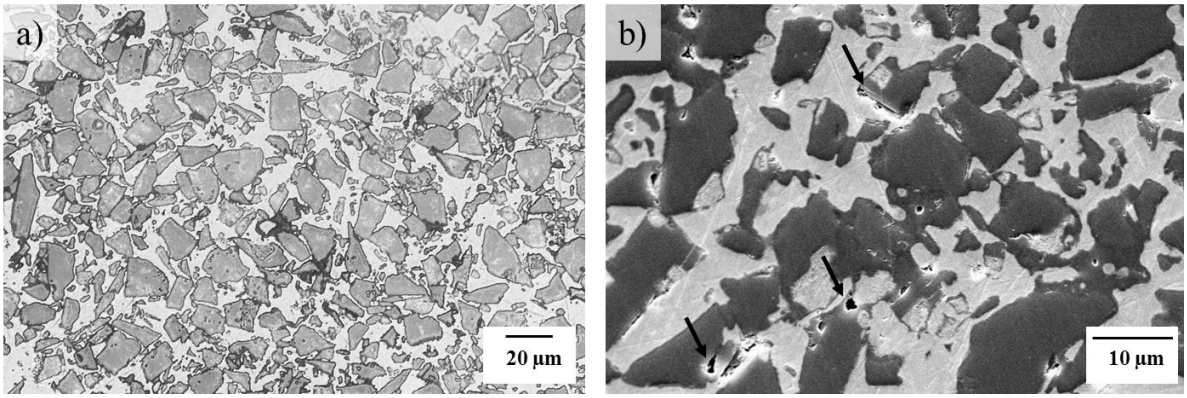

Figure 2. Microstructure of the Cu-SiC-Composite (500x) showing a homogenous distribution of the SiC-Phase inside the composite and porosity at the Cu/SiC-Interface (SEM, 5000x)

Fig. 4 displays the average values of Young's Modulus and Shear Modulus calculated by wave propagation measurements during UPS measurements. These measurements were performed along all three spatial axes of the samples and for each direction three measurements of v_L(longitudinal wave propagation velocity) and v_T (transverse wave propagation velocity) were carried out and were used to determine the elastic constants [17].

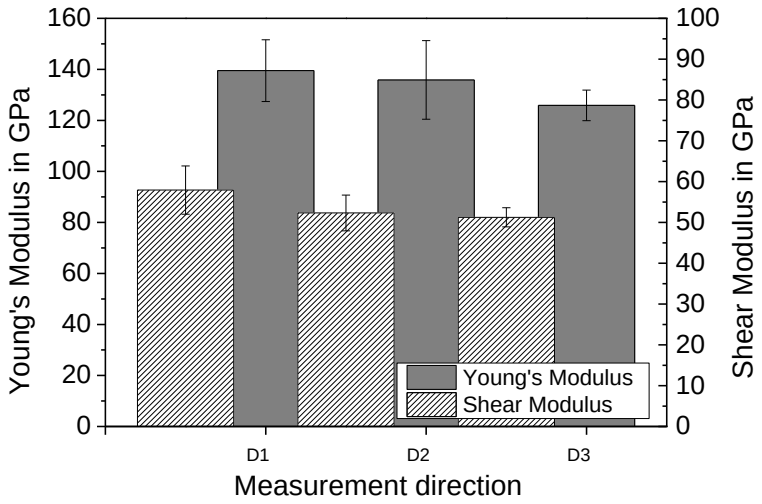

Figure 3. Measured Young's Modulus (E) and Shear Modulus (G) (Directions: D1 = direction of infiltration, D2+D3 = perpendicular to infiltration direction)

Maxima of Youngs-Modulus (E) and Shear Modulus (G) were found in direction D1 (direction of infiltration) with an average of E_{D1} = 139.5 ± 12 GPa and G_{D1}= 57.9 ± 6 GPa. Directions D2 and D3

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(perpendicular to direction of infiltration) showed an average of E_{D2}= 135.9 ± 15 GPa, G_{D2} = 52.3 ± 4 GPa and E_{D3} = 125.9 ± 6 GPa, G_{D3} = 51.2 ± 2 GPa. These values attest a slight anisotropy in elastic constants.

Fig. 4 shows exemplarily stress-strain-diagramms of the composite and the copper matrix material.The compressive strength of the composite is on average of 460 ± 83 MPa . At a total compressive strain of 0.3 %, plastification can be observed. After reaching maximum stress, at 0.95 % compressive strain, a significant decrease of nominal stress occurs resulting from specimen failure by macroscopic crack propagation and fragmentation of the specimen.

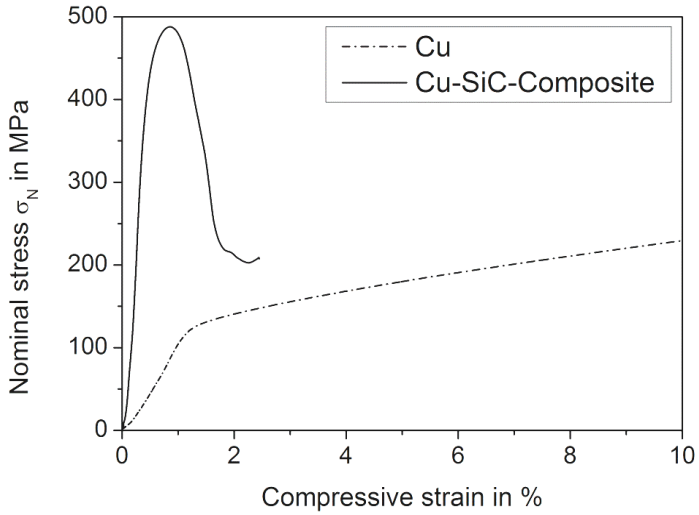

Figure 4. Exemplary stress-compression strain diagrams of Cu-SiC composite samples and Cu-samples

In contrast the copper matrix shows a very ductile behavior and therefore no distinct compression strength. For that reason compression yield strengths $\sigma_{CY0.2}$ and $\sigma_{YC0.5}$ were determined. An overview of the determindend strength values is given in Table 2.

Table 2 Average values for compression strength and compression yield strength determined by quasi-static compression tests

	σ_{UCS} [MPa]	$\sigma_{CY0.2}$ [MPa]	$\sigma_{CY0.5}$ [MPa]
Copper-Matrix	-	116 ± 14	145 ± 15
Composite (D1)	455 ± 93	372 ± 16	454 ± 38
Composte (D2)	466 ± 115	418 ± 76	407 ± 100
Composite (D3)	458 ± 39	428 ± 95	428 ± 110
Composite (Average)	460 ± 83	406 ± 62	429 ± 92

As it can be seen, the composite shows a 0.2% compressive yield strength on average of 406 ± 62 MPa, which is significantly higher than the yield strength of pure copper, which was determined to 116 ± 14 MPa (Increase: + 250 %). The 0.5 % compressive yield strength still offers an increase of 190 %, although the composite specimen is already highly damaged at this stage (confer with Fig. 4 and Fig. 5). There is no significant anisotropy of the strength values found. All values show a high standard variation, which can be ascribed to varation of the porosity fraction and distribution in each specimen. Fig.5 shows an exemplary stress-strain-diagram from an in situ SEM test. Fig.6 shows the associated SEM images taken during the test. Prior to pure elastic deformation, setting effects due to not perfectly parallel surfaces of the specimen can be observed. During elastic deformation (Fig. 5a,b), no structural changes on the specimen surface were observed. Damage initiation was first detected at a compressive strain of $\varepsilon_{c,t}$ = 4.1 % and is accompanied by a small decrease in nominal stress (Fig. 5c).

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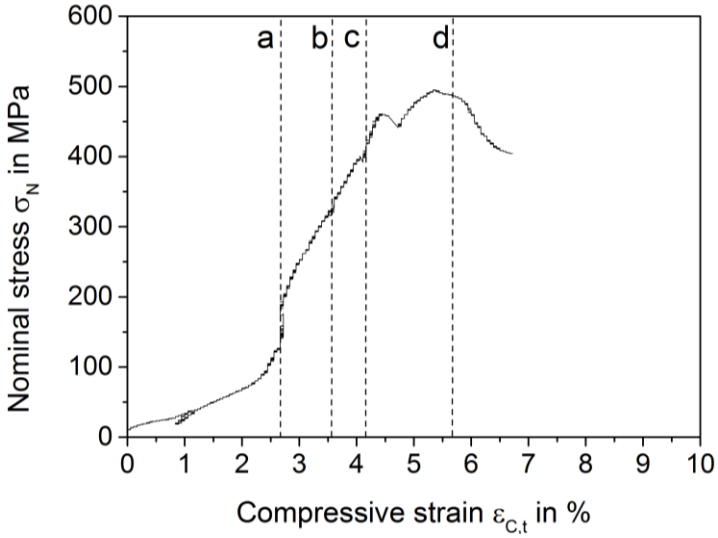

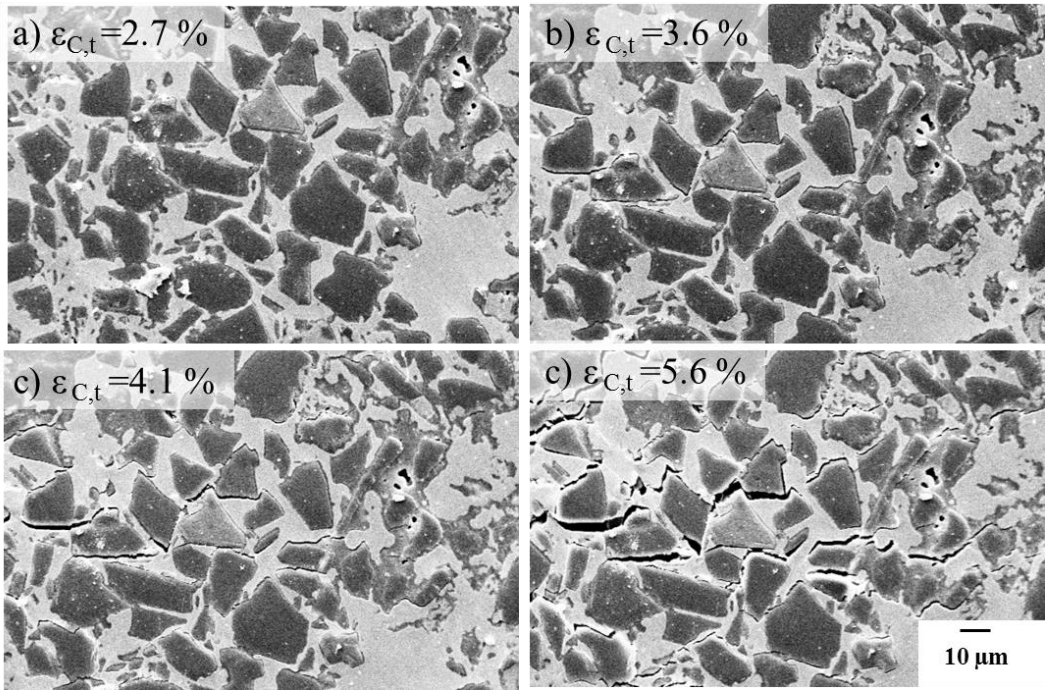

Figure 5 Stress-compression diagram taken during the in situ compression test in the SEM (a-d: surface analysis)

The observation of the specimen surface showed crack initiation along the Cu/SiC-interfaces (Fig. 6c). Most of this interface cracks show an orientation in loading direction, while interfaces perpendicular to loading direction show no visible delamination. Further loading leads to continuous crack growth with a merging of the observed interface cracks. Crack growth and merging than propagates through the metallic phase, leading to the formation of makroscopic cracks (Fig. 6d). Several macroscopic cracks are formed in the specimen, which were all orientated parallel to loading direction eventually passing trough the whole specimen. Further deformation is then characterized by sliding of the resulting fragments.

3. Discussion

Using theoretical values for Young's Modulus of SiC (E_{SiC} = 350 GPa [18]) and measured E for copper (E_{Cu} = 116 GPa) the upper (Voigt [19]) and lower boundaries (Reuss [20]) are calculated to E_V = 253 GPa and E_R = 190 GPa. All measured values lie clearly below the theoretical lower

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boundary. This can be attributed to the high porosity in the composite, which is neglected in the Reuss-Modell and to the formation of SiO_2-layers on the SiC-network during preform synthesis, as described in [16]. This layer is build up to establish sufficient strength of the preforms and can result in SiO_2 fractions of up to 14 vol.-% [16]. For further considerations, it is assumed that the porosity is only present in the Cu-phase and SiO_2-formation reduces only the ceramic phase. If the SiO_2-fraction is then considered with the maximum of 14 vol.-% (with E_{SiO2} = 73 GPa [21]) and the porosity is taken into account by using a model for closed cell foams (described in [22]), the theoretical lower boundary of the Reuss-Modell is reduced to E_R^*= 105 GPa. The upper boundary described by the Voigt-Modell then results in E_V^*= 177 GPa. Consequently the Hill-Modell [23], which showed good coincidence for IPCs in former work [14][15], gives a theoretical Young's Modulus of E_H^* = 141 MPa, which attests a good agreement to the measured values. The damage behavior of IPC's is often characterized by plastic deformation and crack initiation in the metallic phase, e.g. Roy et. al [15] describes the damage accumulation in an IPC with AlSi12-matrix and Al_2O_3-reinforcing structure by crack initiation in the metallic phase due to an arising multi-axial stress state in the matrix [15]. The interface bonding was therefore assumed to be good. In the investigated material system the damage accumulation is obviously characterized by interface failure and subsequent delamination with crack initiation. This indicates a low interface strength, and contradicts the assumption by [16] of a strong interconnected interface in this composite system. As no interfacial reaction is observed, which was required, as interfacial reaction products rather decrease the interfacial strength than improves it [11], the relatively poor wetting behavior and therefore the resulting microporosity at the interface is assumed to be mainly responsible for the poor interface bonding.

3. Summary and Conclusion

A first characterization of a Cu/SiC-composite with interpenetrating network structure manufactured by high temperature squeeze casting of SiC- preforms was performed.

- According to the microstructural investigations a homogenous distribution of the SiC-phase could be achieved. Still a high porosity level of about 15,4 vol.-% was observed.
- The elastic properties were measured by ultrasonic phase spectroscopy. The elastic properties were measured in all specimen directions and showed highest values of Young's Modulus (E) and Shear Modulus (G) in direction of infiltration. Comparison with the Voigt- and Reuss-Modell showed that the measured values lie significantly below the lower boundaries of the theoretical model. This was attributet to the formation of SiO_2-layers on the SiC phase and from the microprosity. Taking into account the maximum volume fraction of SiO_2 and the porosity, a better approximation is achieved. Best approximation is then given by the Hill-Modell.
- The elastic-plastic deformation and damage behavior was studied in compression tests and in situ compression tests. The composite specimens show a significant increase in compression strength properties, but a brittle failure of the specimen after reaching maximum stress. In situ SEM compression tests showed that damage initiation occurs at Cu/SiC-interfaces aligned in loading direction. These original formed cracks lead to further crack propagation and crack merging leading to macroscopic cracks and specimen failure.

For better mechanical performance of this composite system further efforts in preform design (decrease of possible SiO_2-layers) and further development of the HTSC-process (higher process temperature or higher pressure for reducing porosity) have to be the focus of future work.

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