JOURNAL OF MATERIALS SCIENCE **37** (2002) 2575–2579

Wettability and interfacial reaction in SiC/Ni plus Ti system

SHUJIE LI*, YING ZHOU, HUIPING DUAN
Beijing University of Aeronautics and Astronautics, Department of Materials Science and Engineering, Beijing 100083, People's Republic of China
E-mail: shujieli@publica.bj.cninfo.net

Wettability and interfacial reactions in both SiC ceramic/pure Ni system and SiC ceramic/Ni plus Ti system were studied. The contact angle was determined by high temperature photography. The microstructure and composition of the interfacial region were analyzed. Under the applied experimental conditions, the contact angle of SiC/pure Ni system at 1350°C in vacuum is about 86°. The contact angle slightly decreases with the prolonging of time of the test at 1350°C. The interfacial reactions take place during the test. The reactions facilitate the wetting. The wettability of the system is improved by adding active element titanium into the metallic phase. The contact angle decreases with the increasing of content of the additive within the tested range. Moreover, this effect becomes more remarkable with the prolonging of reaction time at high temperatures. This can be explained by the fact that titanium accumulates at the wetting interface and facilitates the interfacial reactions, resulting in an increase of the driving force of wetting.

1. Introduction

The wettability of ceramics by liquid metals is a key factor in many fields of high temperature materials science, especially in metal-matrix composites processing [1] and in metal-ceramic or ceramic-ceramic joining by brazing alloys [2]. In both cases good wettability is generally desired, as it results in large work of adhesion [3] and consequently high interface bonding strength at the metal/ceramic interfaces.

Ni-based superalloys are widely used to manufacture the parts of aeronautical turbine serving at elevated temperatures. The working temperature of these materials can reach 950°C (type GH128) or higher. But it is still not high enough for the next new turbines. In order to further increase the working temperature and decrease the weight of the turbines, new high temperature structural materials are being expected and investigated. Among them, SiC ceramic and SiC matrix composite are promising ones due to their good mechanical strength and resistance to oxidation at elevated temperatures. However, in most cases the application of ceramic materials normally depends on the joining of ceramics to metallic structures [4]. Hence, it is essential to study the wettability of SiC by liquid metals.

The wettability of SiC by liquid pure Ni was reported in Ref. [5], in which reaction bonded SiC and hot pressed SiC were used. As indicated in this reference, the contact angle of the system is about between 65° and 75° at 1773 K in a vacuum, also the contact angle is affected by the kind of SiC used and remarkably affected by the dissolution reaction of Si and C into the liquid metal and carbide formation reaction at the interface between liquid metal and SiC [5]. In Ref. [6], the contact angle of β-SiC/Ni system at 1770 K in a purified argon atmosphere is reported to be 74° [6].

In order to contribute to the joining of SiC ceramic to Ni-based superalloy, it is needed to explore the ways to decrease the contact angle of SiC/Ni system. For this purpose, recrystallized SiC was employed in this study. Besides, taking into account that vigorous carbide formation reaction takes place between titanium and SiC [7–10], titanium was chosen as an additive to improve the wettability of the system. The effect of titanium additive on the wettability of the system has been investigated in this paper.

2. Experimental procedures

The commercial recrystallized SiC ceramic with the purity of >99 wt% was used as the starting ceramic material. The SiC ceramic was cut into cylindrical thin pieces with the Size $\phi 10$ mm × 2 mm. The top surface for testing was polished. Then, the SiC pieces were washed in alcohol with an ultrasonic bath for 20 minutes. After drying, the cleaned pieces were stored in a desiccator. Ni powder with the purity of 99.7 wt% and Ti powder with the purity of 99.2 wt% were used as the metallic materials. Their particle size is −200 mesh.

The content of Ti additive in the tested metallic materials is 1 wt%, 3 wt%, 5 wt%, 7 wt%, 9 wt% and 13 wt% respectively. ("1 wt%" means that the metallic material is composed of 99 wt% Ni + 1 wt% Ti.)

---

*Author to whom all correspondence should be addressed.

The powders were mixed in anhydrous alcohol for 20 minutes. After drying, the mixtures were cold compacted to form cylindrical thin compacts in the size $\phi 10$ mm × 1 mm, which were then cut into small pieces of about $\phi 2$ mm × 1 mm. The small piece of compact was put on the polished surface of the SiC ceramic sample. Then the combination was loaded into the horizontal alumina tube of the "Instrument to Determine the Interface Properties at High Temperature in Vacuum". When the vacuum reached 2×10^{-2} Pa, the specimen was heated up to the melting point of the metallic material. Then, keeping the temperature, the time was recorded, the shape of the metallic drop was observed from the screen, and a photograph was taken every minute. After 6 minutes the specimen was cooled down. The contact angles were measured from the photographs [6]. The microstructure and composition of the specimen were analyzed by Scanning Electron Microscope (SEM) and Energy Dispersive X-ray Microanalysis System (EDX).

The same experiment was carried out with SiC/pure Ni system.

3. Results and discussions

3.1. Wettability in SiC/pure Ni system

In SiC/pure Ni system, it was observed that the liquid Ni drop formed at 1350°C, which is 103°C lower than the melting point of pure Ni. This could be attributed to the fact that significant amount of Si and C dissolved in Ni during heating the sample [5], resulting in the decreasing of the melting point of the metallic material according to the phase diagrams of the binary alloys [11]. The contact angle of the system was determined to be about 86°. The time dependence of the contact angle in the system at 1350°C in a vacuum is presented in Fig. 1. This figure indicates that the contact angle slightly decreases with the prolonging of time.

The microstructure of the SiC/pure Ni system tested at 1350°C in a vacuum for 5 minutes is shown in Fig. 2. One can see that an obvious reaction area exists in the interfacial region. The areas A and B as marked in Fig. 2 were observed by SEM at large magnification, of which the microstructures are shown in Figs 3 and 4 respectively. Although both Figs 3 and 4 show the black alternating with white structures, the black and white phases in Fig. 3 are thicker than that in Fig. 4.

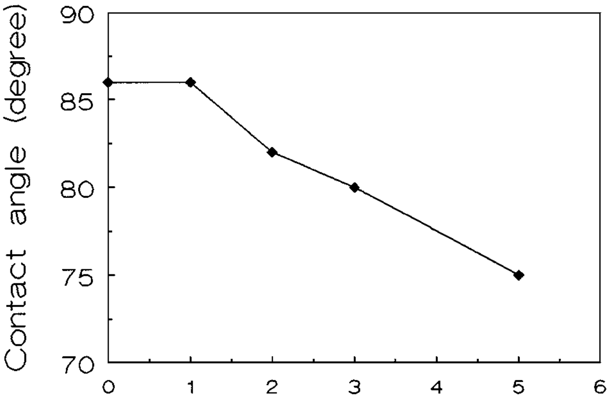

Figure 1 Time dependence of the contact angle in SiC/pure Ni system at 1350°C in vacuum.

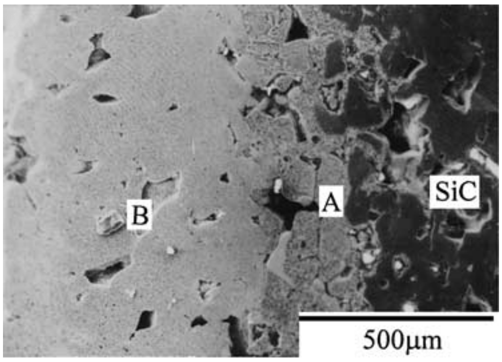

Figure 2 Microstructure of the interfacial region of SiC/pure Ni system tested at 1350°C in vacuum for 5 min.

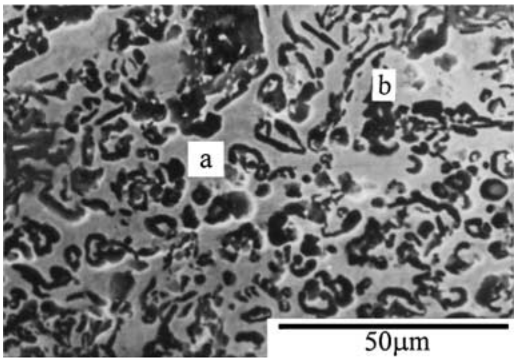

Figure 3 Microstructure of area A as marked in Fig. 2.

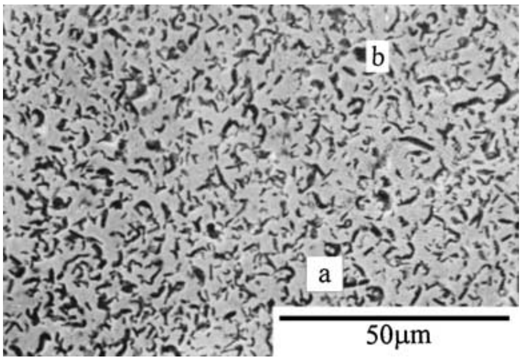

Figure 4 Microstructure of area B as marked in Fig. 2.

The compositions of the white phase denoted by "a" and the black phase denoted by "b" as marked in Figs 3 and 4 were analyzed by EDX. The results are listed in Tables I and II. The atomic ratio of Ni and Si (denoted by Ni : Si) for the various phases was calculated, and the values are also listed in the tables. The Ni : Si for the white phase in Fig. 3 (see Table I, a) is very close

TABLE I Spot analysis results (at%) by EDX at the positions indicated in Fig. 3

Element	a	b
Ni	67.66	8.82
Si	32.34	4.78
C	–	86.4
Ni : Si	2.09	1.85

TABLE II Spot analysis results (at%) by EDX at the positions indicated in Fig. 4

Element	a	b
Ni	71.21	5.98
Si	28.79	1.92
C	–	92.1
Ni : Si	2.47	3.11

to 2, implying that this phase is mainly composed of Ni_2Si. The Ni : Si for the black phase in Fig. 3 (see Table I, b) is also close to 2. Meanwhile, taking into notice the high carbon content of this phase, it is implied that this phase is composed of a mixture of graphite as a major component and Ni_2Si as a minor component. This result is in accordance with that reported in References [12] and [13]. The Ni : Si for the white phase in Fig. 4 (see Table II, a) is close to 2.5, implying the presence of Ni_5Si_2 as the main component. The Ni : Si for the black phase in Fig. 4 (see Table II, b) is close to 3, implying that this phase is composed of Ni_3Si as a minor component and also graphite as a major component. This is in accordance with Reference [14].

The experimental results indicate that diffusion and chemical reactions take place in the wetting test, which make the wetting interface changing from the initial SiC/Ni interface to the reaction products/Ni interface, resulting in the improvement of wettability of the system.

3.2. Wettability in SiC/Ni + Ti system

The contact angles of the SiC/Ni + Ti system with various titanium content at the melting temperatures of the metallic materials are listed in Table III. The melting temperatures are also presented in the table. This table shows that the contact angle decreases gradually with increasing titanium content within the tested range.

Fig. 5 shows the contact angle of SiC/Ni + 5 wt% Ti system as a time function of the test at melting temperature. It is seen that the contact angle decreases remarkably with the prolonging of time, indicating the improvement of wettability. As the work of adhesion at the SiC/Ni + 5 wt% Ti interface is expressed by $W = \sigma_{LV}(1 + \cos\theta)$, where σ_{LV} denotes the surface tension of the liquid alloy, and θ is the contact angle [3], the decrease of θ leads to the increase of W. Therefore, the bonding strength at the interface will be improved by the proper prolonging of time of the test at melting temperature of the metallic material.

TABLE III Contact angle for SiC/Ni + Ti system at melting temperature in vacuum (The specimen was cooled down as soon as the temperature reached the melting point)

Ti content (wt%)	contact angle (°)	Melting temperature (°C)
1	76.6	1380
3	73.7	1442
5	71.5	1333
7	69.9	1290
9	65.2	1290
13	63.5	1340

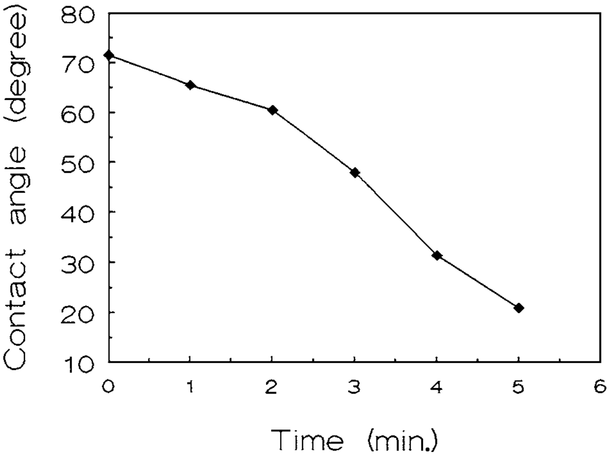

Figure 5 Time dependence of the contact angle for SiC/Ni + 5wt% Ti system at 1333°C in vacuum.

The microstructure and element distribution at the interfacial region of the SiC/Ni + 5 wt% Ti system tested at melting temperature of the metallic material, 1333°C, (The specimen was cooled down as soon as the temperature reached 1333°C.), are presented in Fig. 6. It is seen that the reaction area consists of two layers. The first one, adjacent to SiC, is similar to the reaction area of the SiC/pure Ni system as shown in Fig. 2. The areas A and B in this layer, as marked in Fig. 6, were analyzed by SEM at large magnification and by EDX. Their microstructures are presented in Figs 7 and 8 respectively, also showing the black alternating with white structures. The results of EDX analysis and the calculated Ni : Si for various phases are given in Tables IV and V respectively.

As marked in Figs 7 and 8, "a" denotes the white phase and "b" denotes the black phase. These tables imply that in addition to the other nickel silicides as mentioned above, more Ni_3Si appears in the reaction products, especially in area B. Moreover, the carbon distribution line as shown in Fig. 6 indicates that the carbon content in area B is higher than that in area A.

The second layer, being between the first layer and the remaining metallic drop, is a Ti-rich structure as can

TABLE IV Spot analysis results (at%) by EDX at the positions indicated in Fig. 7

Element	a	b
Ti	–	0.01
Ni	68.27	15.71
Si	31.73	5.21
C	–	79.07
Ni : Si	2.15	3.02

TABLE V Spot analysis results (at%) by EDX at the positions indicated in Fig. 8

Element	a	b
Ti	–	3.03
Ni	75.86	0.07
Si	24.14	0.72
C	–	96.18
Ni : Si	3.14	–

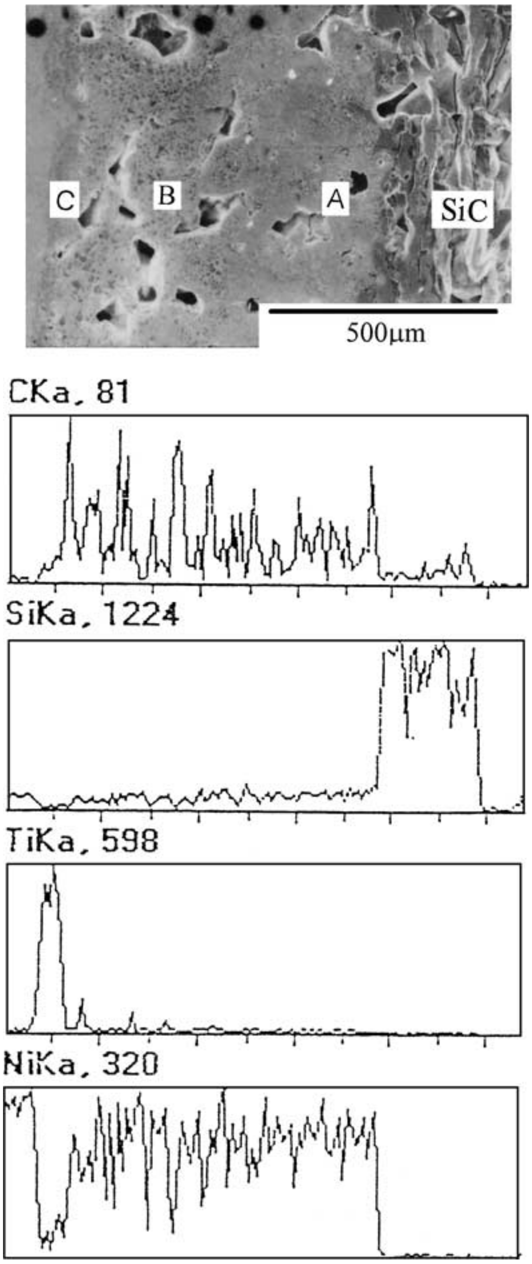

Figure 6 Microstructure and element distribution at the interfacial region of SiC/Ni + 5 wt% Ti system tested at 1333°C in vacuum.

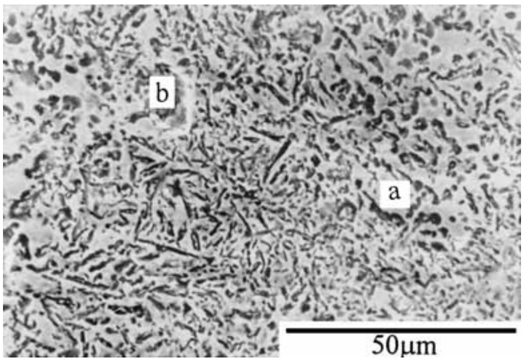

Figure 7 Microstructure of area A as marked in Fig. 6.

be seen from the Ti distribution line in Fig. 6. Area C in this layer, as marked in Fig. 6, was analyzed by SEM at large magnification and by EDX. Its microstructure is presented in Fig. 9. The grey crystals in the figure are Ti-rich phase, which is composed of 98.31 at% Ti and 1.69 at% Ni.

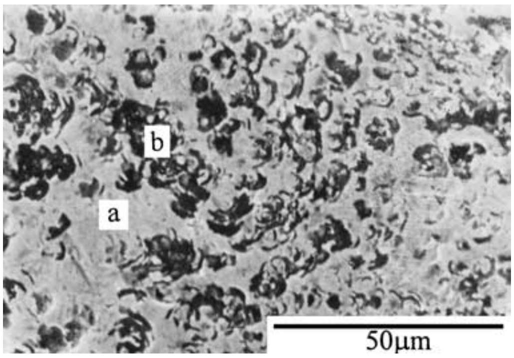

Figure 8 Microstructure of area B as marked in Fig. 6.

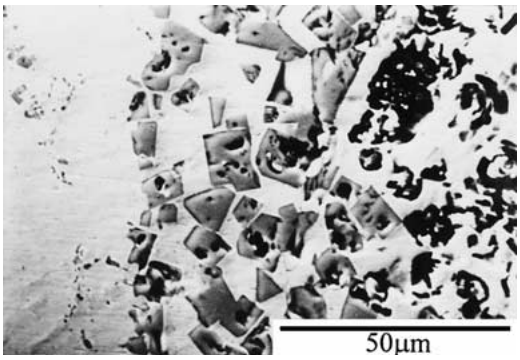

Figure 9 Microstructure of the Ti-rich layer at area C as marked in Fig. 6.

Fig. 10 shows the microstructure and element distribution at the interfacial region of the SiC/Ni + 5 wt% Ti system, tested at 1333°C in a vacuum for 5 minutes. Being similar to that shown in Fig. 6, the reaction area also consists of two layers: a black alternating with white layer and a Ti-rich layer. The element distribution line reveals that the Ti-rich layer is thicker than that in Fig. 6. But its composition does not change significantly, as analyzed by EDX. These results indicate that more titanium accumulates at the wetting interface with the prolonging of time of the test at melting temperature of the metallic material.

As reported, carbon is released as a result of interfacial reaction of SiC and Ti [10]. Also, it is a product of the reaction of SiC and Ni [12–14]. Since titanium is an active element, it has a strong affinity for carbon. This might be the reason for the experimental result that the carbon content of the area near the Ti-rich layer is higher than that of the next area as shown by the carbon distribution lines in Figs 6 and 10, as well as by the data of carbon content in Tables IV and V.

The titanium added to Nickel reacts with SiC during the experiment at high temperatures. Kurokawa and Nagasaki reported the formation of only TiC and Ti_5Si_3 in reaction couples prepared using pressureless sintered SiC and titanium at temperatures between 1173 and 1373 K [15]. Gottselig *et al.* reported that above 1473 K, the primary product was Ti_3SiC_2 [16]. Meanwhile, titanium will react with the graphite, one of the reaction products of SiC and Nickel [12–14], to form stable TiC. This may facilitate the reacting of SiC and Nickel. These interfacial reactions lead to a change of the wetting interface from the initial SiC/Ni + Ti interface to the reaction products/Ni + Ti interface. Besides,

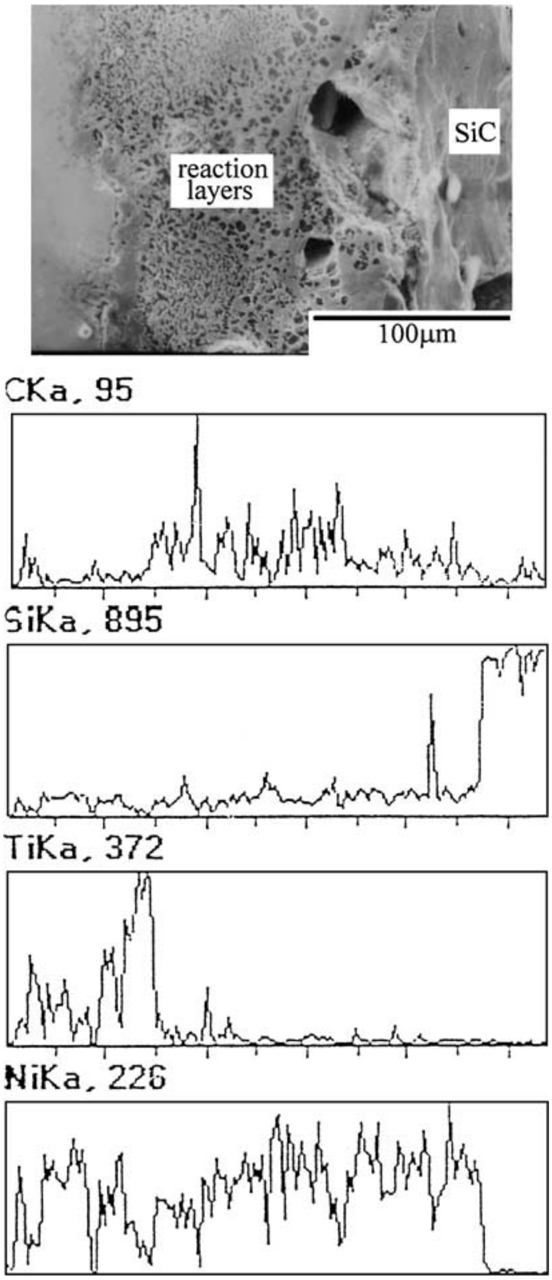

Figure 10 Microstructure and element distribution at the interfacial region of SiC/Ni + 5 wt% Ti tested at 1333°C in vacuum for 5 minutes.

adding titanium to nickel may change the surface tension of the liquid alloy σ_{LV}. These result in the decrease of the contact angle as shown in Table III, which indicates an increase of the driving force of wetting. This can be an explanation of the fact that adding titanium to nickel improves of wettability of the system. This favorable effect becomes more remarkable with the increasing of titanium content and with the prolonging of time of the test at high temperatures, as more reaction products form in these cases.

4. Conclusion

1. Under the experimental conditions of this study, the contact angle of SiC ceramic/pure Ni system at 1350°C in a vacuum is about 86°. The contact angle slightly decreases with the prolonging of time of the test at 1350°C. The chemical reactions take place during the test, indicating that chemical wetting occurs in the system. The reaction products include Ni_2Si, Ni_5Si_2, Ni_3Si and graphite. These reactions facilitate the wetting.

2. Adding titanium to nickel improves the wettability of the system. Within the range tested in this study, the contact angle decreases with the increasing of titanium content and remarkably decreases with the prolonging of time of the test at melting temperature of the metallic material. This can be explained by the fact that active element titanium accumulates at the wetting interface and facilitates the interfacial reactions, resulting in an increase of the driving force of wetting.

3. Under the conditions as described in this paper, appropriate prolonging of time of the test at melting temperatures of the metallic materials will contribute to increasing of the work of adhesion W and consequently the bonding strength at the SiC/Ni + Ti interface.

Acknowledgment

The authors gratefully acknowledge the financial support of the project by the Aeronautical Science Foundation of China (Project No.00H51012) and the National Natural Science Foundation of China (Grant No. 50071002).

References

1. A. MORTENSEN, *Mater. Sci. Engng.* A **135** (1991) 1.
2. M. G. NICHOLAS, *Br. Ceram. Trans. J.* **85** (1986) 144.
3. B. DREVET, S. KALOGEROPOULOU and N. EUSTATHOPOULOS, *Acta Metall. Mater.* **41**(11) (1993) 3119.
4. M. G. NICHOLAS, in "Designing Interfaces for Technological Applications: Ceramic-Ceramic, Ceramic-Metal Joining," edited by S. D. Peteves (Elsevier Applied Science, 1991) p. 49.
5. K. NOGI and K. OGINO, *Transactions of the Japan Institute of Metals* **29**(9) (1988) 742.
6. P. NIKOLOPOULOS, S. AGATHOPOULOS and G. N. ANGELOPOULOS, *J. Mater. Sci.* **27** (1992) 139.
7. S. K. CHOI, M. CHANDRASEKARAN and M. J. BRABERS, *ibid.* **25** (1990) 1957.
8. S. GORSSE and Y. LE PETITCORPS, *Composites Part A* **29A** (1998) 1221.
9. P. MARTINEAU *et al.*, *J. Mater. Sci.* **19** (1984) 2749.
10. M. B. CHAMBERLAIN, *Thin Solid Films* **72** (1980) 305.
11. JIA-XIANG CHEN, "Handbook of Figures. Tables and Data in Common Use for Steel Making" (Metallurgical Industry Press, Beijing, 1984) p. 92 and 98.
12. M. R. JACKSON, R. L. MEHAN, A. M. DAVIS and E. L. HALL, *Metallurgical Transactions* A **14A** (1983) 355.
13. S. K. CHOI, L. FROYEN and M. J. BRABERS, "Joining Ceramics, Glass and Metal," edited by W. Kraft (DGM Informationsgesellschaft, Oberursel, Germany, 1989) p. 297.
14. R. C. J. SCHIEPERS *et al.*, in "Metal-Ceramic Interfaces," edited by M. Ruhle *et al.* (Pergamon Press, New York, 1989) p. 138.
15. K. KUROKAWA and R. NAGASAKI, in "Sintering '87," edited by S. Somiya *et al.* (Elsevier, New York, 1988) p.1397.
16. B. GOTTSELIG *et al.*, "Contribution to the Joining Technique of SiC-Ceramic Using Metallic Interlayers," Juel-2288, Berichte der Kernforschungsanlage, Institut fur Reaktorwerkstoffe, Julich, GmbH, July 1989.

Received 22 August 2000
and accepted 13 February 2002