Ceramics International 23 (1997) 115–118
© 1997 Elsevier Science Limited and Techna S.r.l.
Printed in Great Britain. All rights reserved
PII: S0272-8842(96)00005-3
0272-8842/97 $17.00+.00

The Formation of Aluminium Nitride–Boron Carbide–Aluminium Composites by Wetting Assisted Infiltration

M. Çirakoğlu,[a] Ç. Toy,[b] A. Tekin[a] & W.D. Scott[c]

[a]Faculty of Chemistry and Metallurgy, Istanbul Technical University, Maslak, Istanbul 80626, Turkey
[b]Marmara Research Centre, Gebze-Kocael, Turkey
[c]Department of Materials Science and Engineering, University of Washington, Seattle WA 98195, USA

(Received 13 September 1995; accepted 13 November 1995)

Abstract: The possibility of producing aluminium matrixed aluminium nitride–boron carbide composites by pressureless wetting assisted infiltration has been shown. Cold isostatically pressed cup-shaped preforms of boron carbide (1–10 wt%) aluminium nitride mixtures were sintered at 1400–1600°C in nitrogen atmosphere and then infiltrated with aluminium under 6.5 kPa argon at 1250°C. Pore diameter in the preforms increased with increase in sintering temperature. Infiltrated samples which were sectioned and polished showed no infiltration front. Higher B_4C contents (4–10%) and sintering temperature (1600°C) resulted in full infiltration. In the sintered preforms AlN was the only phase observed. However, in the infiltrated materials both AlN and Al were observed. © 1997 Elsevier Science Limited and Techna S.r.l.

1 INTRODUCTION

Composites of boron carbide and aluminium offer the attractive combination of high hardness and toughness in a lightweight structure, and have potential for a variety of engineering applications. Aluminium nitride is a hard refractory material and has attractive engineering properties like high thermal conductivity and low thermal expansion coefficient in addition to a low density. It is useful in heat sink applications and in lightweight structural parts. The combination of both aluminium nitride and boron carbide with aluminium results in an enhancement of the properties of the cermet than when only either of the ceramics is used. There are several methods which have been used for manufacturing metal-matrix composites such as these. One of them is pressureless wetting assisted infiltration.[1–3] In this method, molten metal wets the ceramic and fills the pores throughout the body. Wetting assisted infiltration without the application of pressure has advantages for the production of complex shaped parts to near net shape with low residual porosity.

The wettability of a solid by a liquid is indicated by the contact angle between the two phases. If the liquid drop spreads completely over the solid surface, contact angle becomes zero, and ideal wetting occurs. In a non-wetting system[1] contact angle is 180°. The wetting of ceramic surfaces by molten metals is critical for the processing of AlN/Al composites if a good bending strength is desired from the final composite.[2] Chemical reactions between liquid and solid surfaces can result in good wetting. This is easily achieved in metallic systems but it is not easy in a metal–ceramic system. Even though chemical reactions enhance wettability and infiltration, reaction products may disrupt microstucture and lower strength.[3] However, the chemical reaction between B_4C and Al results in good wetting and increased infiltration.[4] The wettability of AlN by Al increases with increasing temperature and wetting starts above 850°C under vacuum conditions.[2]

2 EXPERIMENTAL

Commercially available AlN (Hermann C. Starck Inc., Berlin, average particle size 13.6 μm, N 33%, C 0.05%, O 0.7% and Fe 0.01%) and B_4C (ESK Inc., particle size 5 μm) were used as the starting materials.

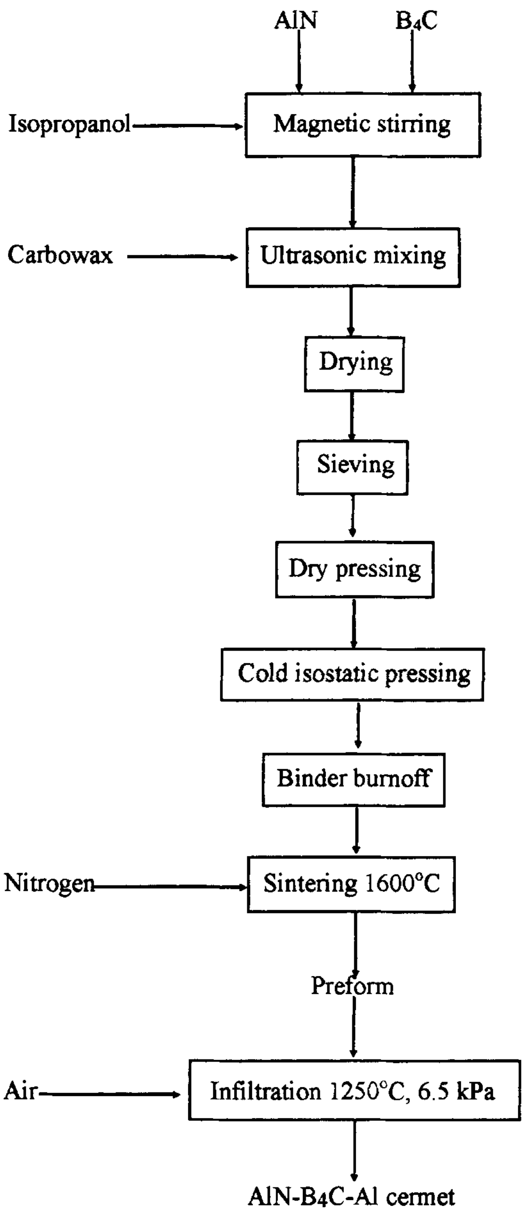

Fig. 1. Flow diagram for the processing of AlN–B_4C–Al composites.

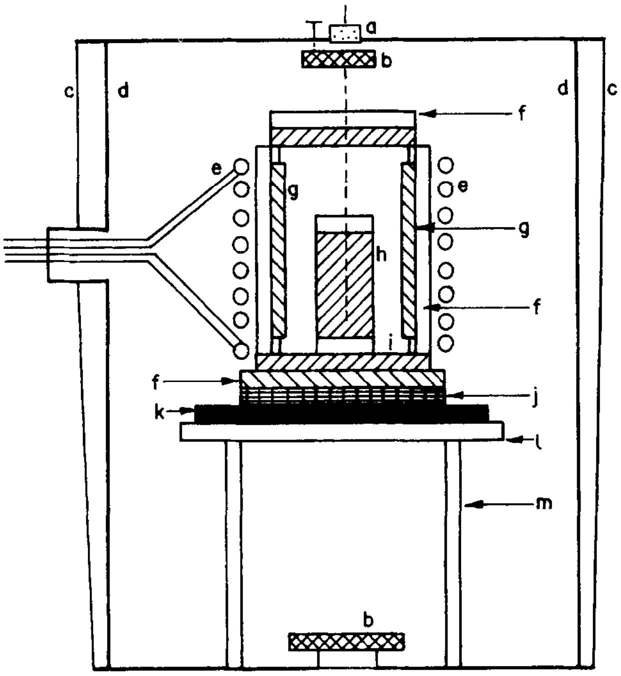

Fig. 2. Schematic picture of the sintering furnace: (a) silica sight glass; (b) Mo radiation shield; (c) water jacket; (d) bronze chamber case; (e) copper tube and induction coil; (f) graphite felt; (g) graphite susceptor; (h) graphite crucible with top and bottom; (i) graphite step; (j) honeycomb cordierite insulator; (k) SiC plate; (l) steel plate; (m) steel stand.

The sequence of steps in the experiments is indicated in the flow diagram given in Fig. 1. Aluminium nitride powder was mixed with calculated quantities of boron carbide powder, corresponding to B_4C contents of 1, 2, 4 and 10 wt% in the mixture. Since AlN reacts with water, the mixture was prepared in 2 propanol at room temperature using a magnetic stirrer, followed by ultrasonic agitation. During ultrasonic mixing, 4 wt% binder (carbowax, polyethylene glycol obtained from Union Carbide) was added to give green strength while handling. Then the suspension was dried at 60°C to evaporate all alcohol and the dried mixture was sieved through an ASTM No.20 mesh sieve. The powder thus obtained was weighed and uniaxially pressed at 28 MPa in a steel die to form cup-shaped specimens. These specimens were then cold isostatic pressed at 172 MPa. Binder was burnt off by heating in air at 1°C/min to 200°C and keeping at this temperature for 1 h. Specimens were placed on AlN powder in a graphite crucible shown schematically in Fig. 2, and sintered under nitrogen atmosphere at 1400, 1500 and 1600°C. Heating rate was about 12°C/min and holding time 1 h. Nitrogen was dried by passing through drierite and purified by passing through copper turnings at 350°C before entering the furnace. The same induction furnace was used for infiltration experiments. The sintered specimens were placed on AlN bedding powder in contact with a known amount of aluminium (1100 grade Al) metal. The specimens were heated under vacuum to 900°C and argon was then admitted into the furnace. Argon was purified like nitrogen. Additionally, the inert gas also passed through a titanium sponge containing furnace at 800°C. The pressure of argon inside the furnace was maintained at 6.5 kPa through partial evacuation.

Mercury porosimetry was used to characterize the preform structure. Infiltrated samples were sectioned through the diameter with a diamond saw to determine infiltration depths. Phase identification in the preform and infiltrated samples was done by X-ray diffraction. Infiltrated samples were also examined by optical microscopy.

Table 1. Pore diameter (nm) change with sintering temperature and composition

	Temperature (°C)	
Sample	1400	1600
AlN+10% B$_4$C	112	631
AlN+4% B$_4$C	142	686
AlN+1% B$_4$C	145	1121

3 RESULTS AND DISCUSSION

The pore diameter in the composite preforms as a function of temperature and boron carbide content are given in Table 1. The pore diameter increases with an increase in sintering temperature. However, as the B$_4$C content in the mixture increases the pore diameter decreases. This may be related to the smaller particle size of B$_4$C.

Experimental results[5–7] in the infiltration of liquid metals in porous preforms qualitatively illustrate that the rate of infiltration is directly proportional to pore diameter, time and surface energy of the liquid phase. Present results in the B$_4$C–AlN–Al system also indicate that the pore diameter is not the only factor which affects infiltration. Even though pore diameter in AlN + 1%B$_4$C samples (145 nm) is larger than that in the AlN + 10% B$_4$C samples (112 nm), infiltration did not take place in the former. The presence of a thermodynamically stable oxide layer (like Al$_2$O$_3$) on the preforms will inhibit the wetting and infiltration. The good infiltration at higher B$_4$C content suggests that the oxide layer undergoes reduction and removal by reducing agent B$_4$C.

The sections through the infiltrated samples revealed no infiltration front. In the specimens used, infiltration can take place from the top where aluminium initially contacts the preform, as well as from the sides towards the centre of the sample as liquid aluminium flows and wets the sample side surfaces.

New compounds (like BN) formed by chemical reactions during sintering could enhance infiltration. However, XRD patterns of the sintered preforms revealed no new phases. The pattern is shown in Fig. 3. The XRD pattern of the infiltrated material (AlN + 4% B$_4$C) is shown in Fig. 4. It indicates AlN and Al as main phases.

Examination of the infiltrated samples in the optical microscope showed that the infiltrated aluminium surrounded the AlN and B$_4$C grains resulting in an aluminium matrix composite as shown in Fig. 5.

4 CONCLUSIONS

The formation of AlN–B$_4$C–Al composites by wetting assisted infiltration can be accomplished by a two-step process. The first step is the formation of AlN–B$_4$C, a ceramic perform having continuous open porosity, by cold pressing of the ceramic powder mixture followed by sintering at 1600°C under nitrogen. The second step is infiltration with molten aluminium metal at 1250°C under 6.5 kPa argon.

The B$_4$C content in the composite should be at least 4 wt% and the perform sintering temperature

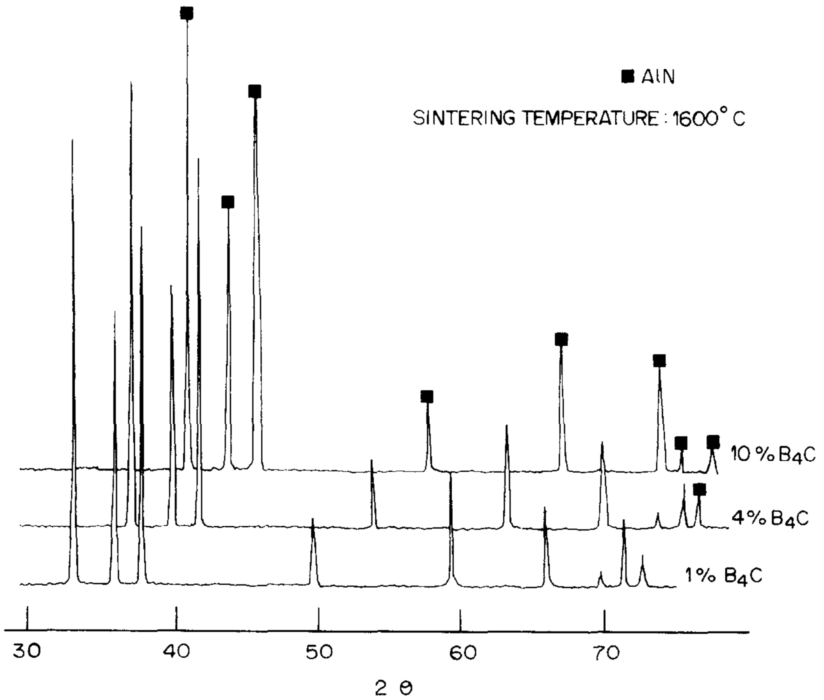

Fig. 3. XRD pattern of sintered preforms.

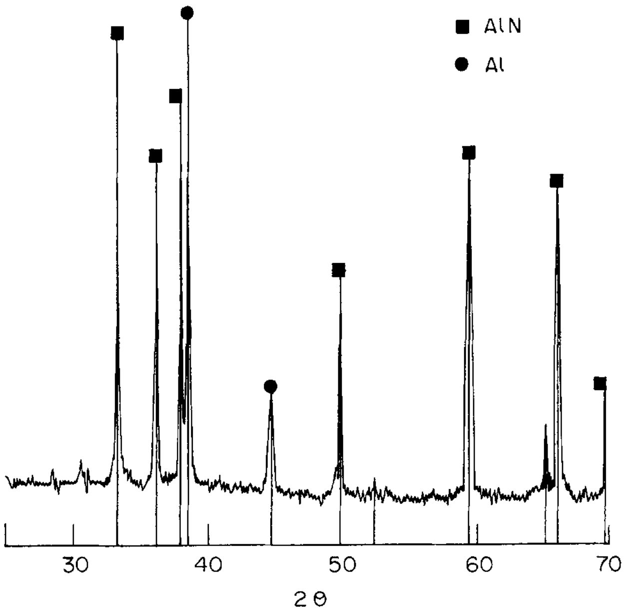

Fig. 4. XRD pattern of infiltrated samples.

must be at least 1600°C for achieving full infiltration. If the B_4C content is 10 wt%, it is possible to provide infiltration in samples which are sintered at 1400°C and 1600°C. Enhanced infiltration with higher B_4C content in the composite could result from the reducing action of the carbide which supresses the formation of a stable wetting inhibiting oxide layer on the preform.

ACKNOWLEDGEMENT

The authors thank the United Nations Industrial Development Organization (UNIDO) for financial support.

Fig. 5. Optical micrograph of infiltrated sample ($AlN+4\%B_4C$).

REFERENCES

1. BANERJI, A., ROHATGI, P. K. & REIF, W., Role of wettability in the preparation of metal-matrix composites: A review. *Metall.*, **38** (1984) 656–61.
2. TOY, C. & SCOTT, W. D., Ceramic–metal composite produced by melt infiltration. *J. Am. Ceram. Soc.*, **73**(1) (1990) 97–101.
3. DELANNAY, F., FROYEN, L. & DERUYTTERE, A., The wetting of solids by molten metals and its relation to the preparation of metal matrix composites: A review. *J. Mater. Sci.*, **22** (1987) 1–16.
4. HALVERSON, D. C., PYZIK, A. J. & AKSAY, I. A., Processing and microstructural characterization of B_4C–Al cermets. *Ceram. Eng. Sci. Proc.*, **6** (1985) 736–44.
5. HILLIG, W. B., Ceramic matrix composites by means of melt infiltration. *Ceram. Eng. Sci. Proc.*, **8** (1987) 834–8.
6. SEMLAK, K. A. & RHINES, F. N., The rate of infiltration of metals. *Trans. Metall. Soc. AIME*, **212** (1958) 325–31.
7. KURILKO, A., KURSHEV, G. A., RUDYUK, V. A. & NAIDICH, Y. N., Kinetic laws of infiltration of porous Ti by Pb and in melts. *Poroshk. Metall. (English translation)*, **9**(261) (1984) 35–8.