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Al_2O_3/SiC Platelet Composites. Effect of Sintering Conditions

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Abstract

Al_2O_3/SiC-platelet composites have been obtained by three different sintering routes: pressureless sintering, gas pressure sintering and hot pressing. The effect of sintering conditions on the densification and microstructure of the composites has been evaluated.

For the compacts obtained by hot pressing, relative densities close to theoretical have been achieved at temperatures 200–300°C lower than that used to sinter Al_2O_3/SiC-whisker composites. This technique produced anisotropic composites where the platelet basal plane was slightly perpendicular to the hot pressing axis.

Gas pressure sintering has been proved to be an alternative to hot pressing for developing high-density near-net-shape Al_2O_3/SiC-platelet components with SiC contents up to 12 vol%. © 1996 Elsevier Science Limited.

1 Introduction

During the last decade, the development of ceramic matrix composites, especially SiC whisker-reinforced Al_2O_3 composites, has increased because of their excellent thermomechanical properties which allow their use as cutting tools and as components for structural applications at high temperatures.

Although the best mechanical properties have been achieved when whiskers are incorporated into the alumina matrix,[1,2] recently, the present authors[3] have proved, using double torsion tests, that Al_2O_3 with 30 vol% SiC platelets presented a higher threshold stress intensity factor for slow crack growth ($K_0 \sim 9$ MPa $m^{1/2}$) than that obtained for Al_2O_3 with SiC whiskers ($K_0 \sim 6$ MPa $m^{1/2}$). Additionally, platelets are now increasingly employed because they are not so dangerous to health as whiskers,[4] and also because of the simplicity of handling and the possibility of introducing higher contents without agglomeration problems.[5]

There are several works devoted to the study of processing[6–8] and mechanical properties[3,8–14] of SiC-platelet (SiC-pl) reinforced Al_2O_3 matrix. In these works, the Al_2O_3/SiC-pl composites have been sintered by hot pressing, a technique that only allows the fabrication of pieces with simple geometry that require extensive machining. On the other hand, hot pressed Al_2O_3/SiC-pl composites have shown anisotropic mechanical and wear behaviour due to the preferred orientation of the platelets with respect to the hot pressing axis.[7,9,15] Consequently, it is important to consider other sintering routes, such as pressureless and gas pressure sintering, which can be used to develop isotropic near-net-shape components with lower final cost.

In the present work, Al_2O_3/SiC-pl composites have been obtained by three different sintering routes: pressureless sintering, gas pressure sintering and hot pressing. The major objective has been to study the effect of processing parameters, such as SiC content, sintering temperature and soaking time, as well as the sintering route (i.e. hot pressing, pressureless and gas pressure sintering) on the densification and the microstructure of Al_2O_3/SiC-platelet composites.

2 Experimental Procedure

A submicronic α-Al_2O_3 with a mean particle size of 0·3 μm (CS400, Lonza Martinswerk, Germany) and SiC platelets with 16 μm mean diameter size (grade SF, C-Axis Technology, Canada) were used to prepare powder/platelet homogeneous suspensions with platelet contents ranging from 0 to 30 vol%. These suspensions were processed by pH variation using the flocculation method described in a previous work.[6] In this way, Al_2O_3/SiC-pl aqueous

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suspensions with 70 wt% solids content were stabilized at pH = 10 and then flocculated by adding HCl up to pH = 7. The viscous flocculated slip was dried at 120°C for 24 h and sieved up to 100 μm.

Pressureless sintering, gas pressure sintering and hot pressing were employed to study the effect of the sintering route on densification and microstructure of Al_2O_3/SiC-pl composites. For pressureless and gas pressure sintering, the powder/platelet green compacts were prepared by isostatic pressing at 200 MPa.

2.1 Pressureless sintering

Pressureless sintering (PS) treatments of compacts containing up to 12 vol% of SiC platelets were performed in argon, at temperatures and times ranging between 1650 and 1800°C, and between 1 and 4 h. These treatments were carried out in a vertical furnace with graphite resistances. Nieto *et al.*[16] proved that in Al_2O_3/SiC-particles composites sintered under similar conditions, i.e. in a graphite furnace at 1700°C and using Ar as sintering atmosphere, a reduced layer containing metallic Al and Si particles was formed during sintering. To avoid that reduction process, the samples were introduced in a SiC powder bed.[16]

2.2 Gas pressure sintering

Gas pressure sintering (GPS) consisted basically of two steps. The first, in which a close pore stage was reached, involved densification under a low gas pressure (1 MPa) up to the holding temperature. In the second step, a high external gas pressure was introduced to promote pore elimination.

GPS treatments were performed on samples with SiC platelet content ranging between 0 and 30 vol%. Two different sintering conditions were used:

(i) T1: 1760°C for 1 h 45 min applying 7 MPa of nitrogen pressure. These treatments were performed in an industrial furnace which had a limiting gas pressure of 10 MPa.
(ii) T2: 1650°C during 30 min with an argon pressure of 17 MPa. In this case, the GPS treatments were made in an autoclave that allowed the use of gas pressures higher than 10 MPa but lower temperatures.

2.3 Hot pressing

Hot pressed discs (HP) were obtained at 50 MPa of applied pressure in argon atmosphere at 1500°C during 30 min for SiC-pl contents <12 vol% and at 1550°C during 1 h for contents ≥ 12 vol%. Platelet orientation during hot pressing was quantified by determining both the platelet aspect ratio and the area filled by the SiC platelets in two different planes of the specimen, which corresponded to the surfaces parallel and perpendicular to the hot pressing direction. This analysis was performed, as a function of platelet content, on optical micrographs using an automatic image analyser. At least 200 features were considered in each analysis.

Final densities were measured in water by the Archimedes method. The formation of reaction products during sintering was studied by X-ray diffraction (XRD). The microstructure of the samples was analysed on polished surfaces using optical microscopy (OM).

3 Results and Discussion

3.1. Densification

3.1.1. Pressureless sintering

Relative density as a function of pressureless sintering temperature of Al_2O_3/SiC-platelet composites is plotted in Fig. 1. The results revealed that the highest densities, 96 d_{th}% for the composite with 5 vol% of SiC-pl and 85 d_{th}% for that with 12 vol% of SiC-pl, were achieved at 1700 and 1750°C, respectively. At 1700°C, the relative density decreased for holding times longer than 2 h up to 90 d_{th}% for the specimen with 5 vol% of SiC-pl, and 80 d_{th}% for that containing 12 vol% of SiC-pl.

Densification rate showed a strong decrease with platelet content, as corresponds to the sintering behaviour of heterogeneous ceramic powders which contain non-sinterable inclusions.[17–20] In this case, matrix regions of compression are developed around interactive platelets which densify

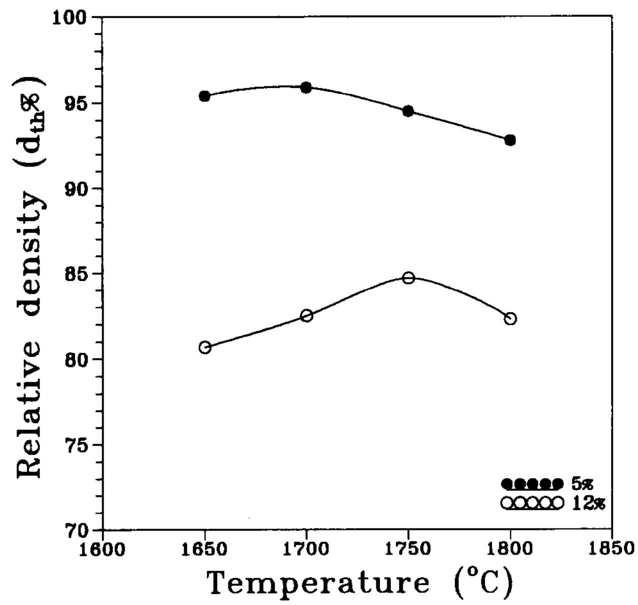

Fig. 1. Relative density of pressureless sintered Al_2O_3/SiC-pl composites containing 5 and 12 vol% of SiC platelets as a function of sintering temperature at a holding time of 2 h.

Table 1. Weight loss (wt %) and relative density (d_{th} %) of Al_2O_3/SiC-pl composites after pressureless sintering treatments

Heat treatment		*5 vol% SiC*		*12 vol% SiC*	
Temperature	*Time*	*wt%*	*d_{th}%*	*wt%*	*d_{th}%*
1650°C	2 h	2·6	95·4	2·8	80·7
1700°C	2 h	3·9	95·9	4·2	82·5
1750°C	2 h	5·0	94·5	6·7	84·7
1800°C	2 h	7·1	92·8	10·5	82·3
1700°C	1 h	3·9	92·1	3·3	80·3
	2 h	3·9	95·9	4·2	82·5
	3 h	5·6	93·0	6·1	81·8
	4 h	6·2	90·5	7·3	79·6

first, producing a non-deformable network and constraining the shrinkage of the adjacent porous matrix. This constraint causes desintering and crack-like void formation, enhancing the porosity and decreasing the final density of pressureless sintered Al_2O_3/SiC-pl composites. The magnitude of this effect depends on the non-sinterable particles' content. In fact, for 12 vol% SiC-pl, the density fell dramatically to 82 d_{th}% (Fig. 1).

Weight loss of the samples during the different PS treatments is summarized in Table 1. As can be observed in this table, the weight loss increased with temperature, time and SiC platelet content from 2·6% for the compact with 5 vol% of SiC-pl heat treated at 1650°C/2 h up to 10·5% for 12 vol% SiC-pl-containing composite at 1800°C/2 h. In this last specimen, an external layer was observed which was analysed by XRD.

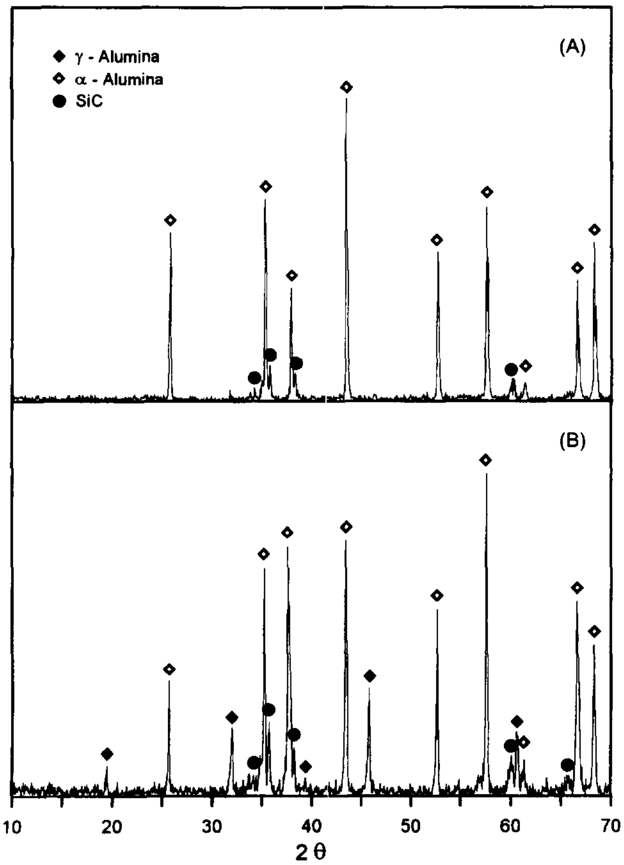

Fig. 2. XRD patterns corresponding to (A) the bulk and (B) the surface of the Al_2O_3/12 vol% SiC-pl compact sintered at 1800°C during 2 h.

According to the XRD patterns of the surface and the bulk of the Al_2O_3/12 vol% SiC-pl sample (Fig. 2), γ-Al_2O_3 was detected at the surface of the specimen while only α-Al_2O_3 and SiC were observed in the bulk. Therefore, the reducing sintering atmosphere led to the formation of an Al_2O_3 structure (γ-Al_2O_3), in which Al ions show lower coordination than in α-Al_2O_3; i.e. α-Al_2O_3 lattice represents a hexagonal closest packing with Al^{3+} octahedrally coordinated while γ-Al_2O_3 presents a defect spinel structure in which aluminum is both octahedrally and tetrahedrally coordinated.

The strong increase in weight loss, observed in samples with high SiC content treated at 1800°C for long times, could be explained by both the reduction of the amorphous SiO_2 always present at the surface of SiC platelets, and by the oxygen loss associated with the formation of γ-Al_2O_3.

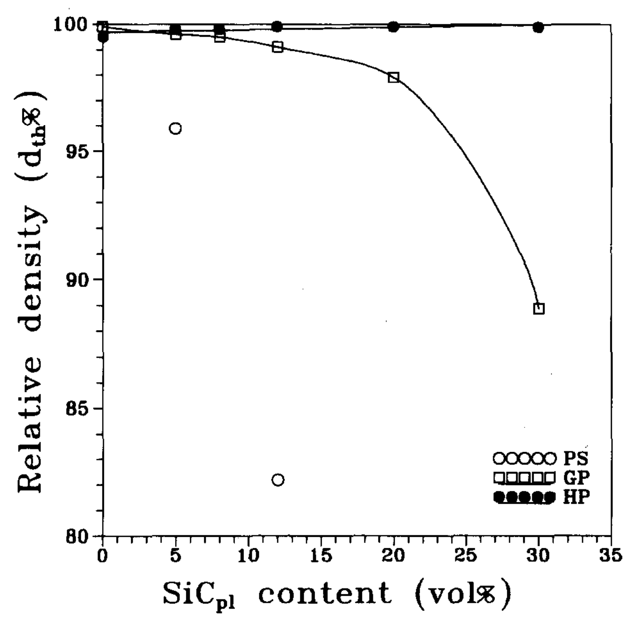

Fig. 3. Relative density as a function of SiC-pl content for Al_2O_3/SiC-pl composites obtained by: (PS) pressureless sintering at 1700°C for 2 h, (GP) gas pressure sintering (T1 treatment) and (HP) hot pressing.

Table 2. Relative densities of Al_2O_3/SiC-pl composites as a function of gas pressure sintering treatments

Treatment	*SiC_{pl} content (vol%)*	*Density (d_{th}%)*
T1	5	99·6
1760°C/1 h 45 min	8	99·5
7 MPa/N_2	12	99·1
	20	97·9
	30	88·9
T2	5	99·4
1650°C/30 min	8	99·0
17 MPa/Ar	12	94·6
	20	86·5

3.1.2 Gas pressure sintering

The employment of a gas pressure during sintering enhanced the final density of SiC-platelet-containing composites compared to that obtained by pressureless sintering, as can be clearly observed in Fig. 3. This behaviour is due to the improvement in sintering driving force and also to the use of a tight sintering atmosphere which will considerably reduce chemical reactions and, therefore, weight loss.

The best GPS results were achieved with T1 treatment, which corresponds to the highest-temperature process, obtaining relative densities higher than 99 d_{th}% for SiC platelet content up to 12 vol% (Table 2). This fact shows that a high temperature (1760°C) is necessary to eliminate open porosity, while a relatively low nitrogen pressure (7 MPa) compared to that required in T2 can be used to increase the final density.

On the other hand, when nitrogen was used as sintering atmosphere, an external reaction layer of ~20 μm was detected over the sample surface. The XRD analysis of this layer revealed the presence of 15R-SiAlON, γ-AlON and AlN formed by a carbothermal reduction of Al_2O_3. Only Al_2O_3 and SiC were detected in the bulk of the sample.

3.1.3 Hot pressing

In the hot pressing technique, the driving force for sintering is increased by applying mechanical energy. This technique allowed fully dense compacts to be obtained for platelet content up to 30 vol% (Fig. 3). For high SiC platelet content (≥ 12 vol%), a higher support of energy was necessary to exceed the backstresses opposite to the densification. In this case, the temperature and the time were enhanced from 1500°C and 0·5 h to 1550°C and 1 h, respectively. These temperatures were about 200–300°C lower that those generally used to densify Al_2O_3/SiC-whiskers composites.

3.2 Microstructure

Optical micrographs corresponding to pressureless and gas pressure sintered compacts with 12 vol% SiC-pl are shown in Fig. 4. Gas pressure sintered material showed a denser microstructure than the pressureless sintered one, virtually without open porosity (Fig. 4A). The samples sintered using both techniques presented pores largely concentrated in regions adjacent to the SiC platelets, especially along the platelet surface, where tensile stresses are formed. At the ends of the platelets and at locations between closely spaced platelets, dense regions were developed due to the presence of compression stresses during sintering. This fact is a consequence of the differential shrinkage rate of Al_2O_3 matrix and SiC platelets.[20]

In Fig. 4, a random orientation of the platelets into the matrix is observed in the materials obtained by pressureless sintering and gas pressure sintering.

In hot pressed Al_2O_3/SiC-pl composites, a microstructure completely free of pores can be observed (Fig. 5). In this figure, a preferred orientation of the platelets was detected. The plot of the area fraction filled by the SiC platelets in the parallel plane versus the perpendicular one (Fig. 6) shows that SiC area was slightly higher in

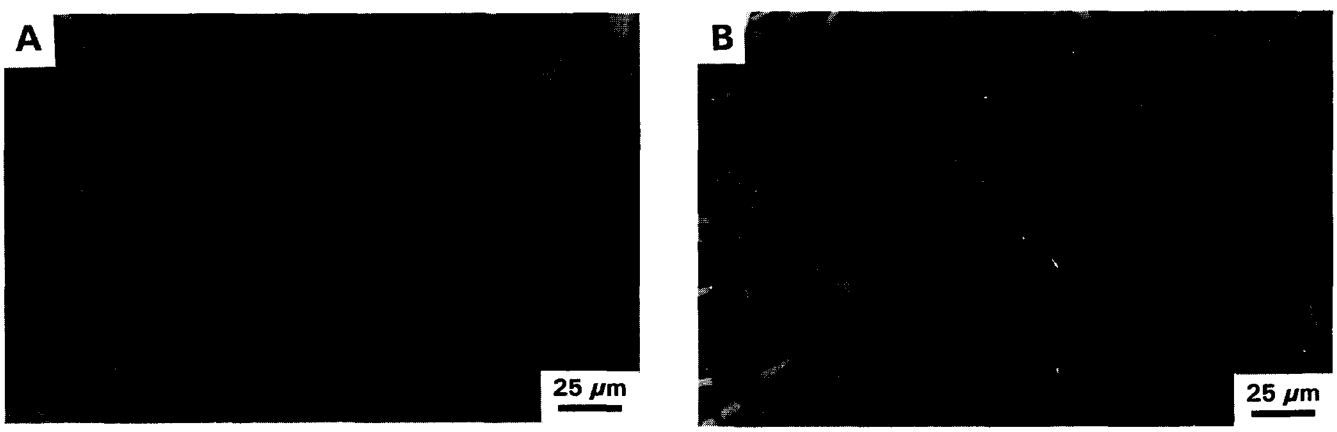

Fig. 4. Optical micrographs of Al_2O_3/12 vol% SiC-pl composites obtained by: (A) gas pressure sintering (treatment T1) and (B) pressureless sintering at 1750°C for 2 h.

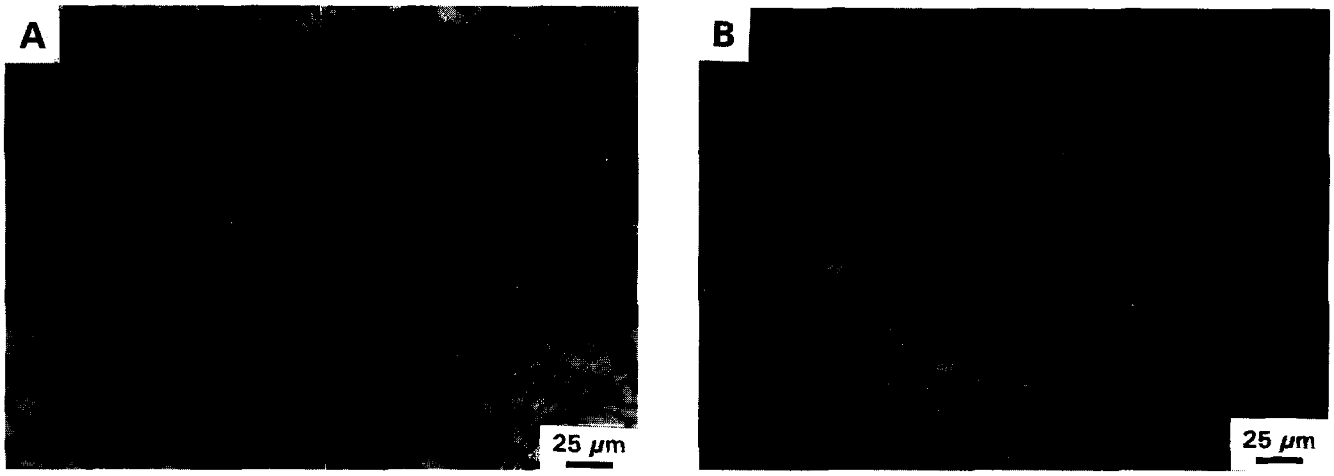

Fig. 5. Optical micrographs of the Al_2O_3 compact with 30 vol% of SiC-pl hot pressed at 1550°C for 1 h, showing the planes (A) perpendicular and (B) parallel to the hot pressing axis.

the perpendicular plane than in the parallel one, which indicates that platelets were oriented with basal planes perpendicular to the hot pressing axis.

On the other hand, the filled area ratio between parallel and perpendicular planes becomes closer to 1 as SiC platelet content increases (Fig. 6) and therefore, the level of platelet orientation decreases with platelet content.

The decrease in platelet orientation with the platelet content was also pointed out by quantifying the platelet aspect ratio in the parallel and perpendicular planes (Fig. 7). For SiC platelet content ≤ 12 vol%, the aspect ratio of the platelets in the parallel plane was higher than in the perpendicular one while, for Al_2O_3/30 vol% SiC-pl compact, this difference in aspect ratio between both planes significantly decreased.

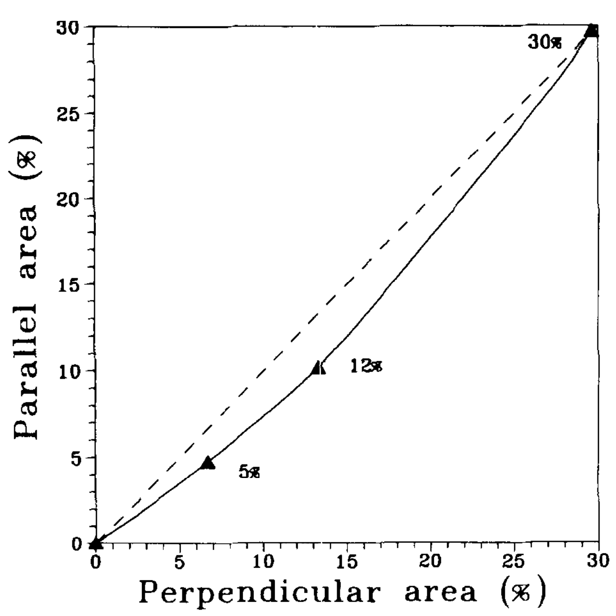

Fig. 6. Filled area (%) occupied by the SiC platelets in the parallel plane versus the perpendicular one, for three different SiC platelet contents.

When SiC platelet contents increase above 30 vol%, platelets percolate forming a rigid skeleton that will impede the platelet orientation during hot pressing.

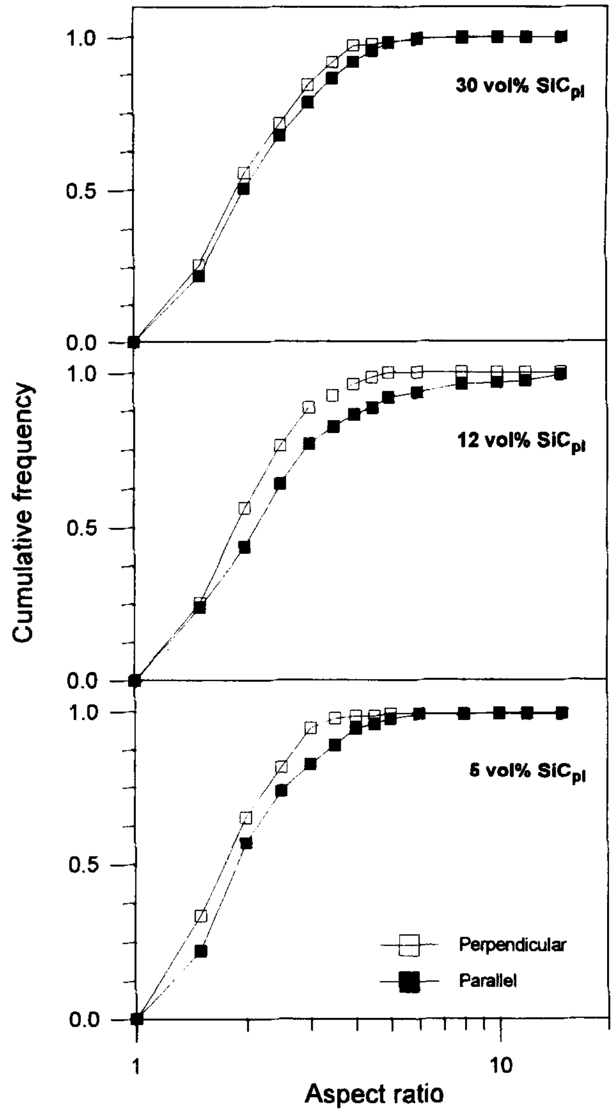

Fig. 7. Platelet aspect ratio distribution in the perpendicular and parallel planes for Al_2O_3/SiC-pl composites containing 5, 12 and 30 vol% of SiC platelets.

4 Conclusions

Many factors affect the densification of Al_2O_3/SiC-platelet composites, the most important being the temperature and soaking time of the heat treatment and, mainly, the SiC platelet content and the sintering route, i.e. pressureless sintering, gas pressure sintering and hot pressing.

The reducing atmosphere, developed in a graphite furnace during sintering in Ar, has led to the formation of γ-Al_2O_3 at the surface of the samples pressureless sintered at 1800°C/2 h.

For SiC platelet contents up to 30 vol%, high density values have been achieved using hot pressing at temperatures 200–300°C lower than that employed to densify Al_2O_3/SiC-whisker composites. During the thermal treatment, platelets tend to a preferred orientation with basal planes perpendicular to the hot pressing axis. The grade of orientation decreased when SiC-pl content increased.

Gas pressure sintering has been found to be a good alternative to hot pressing to produce near-net-shape ceramic components. With this technique, relative densities close to theoretical have been obtained for SiC platelet contents ≤ 12 vol%.

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