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Obtention of highly dispersed platelet-reinforced Al_2O_3 composites

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The rheological behaviour of alumina/platelets (Al_2O_3 or SiC) suspensions has been established by measuring zeta potential versus pH and the relative viscosity as a function of pH, solids loading and platelets content. Highly homogeneous mixtures of alumina/platelets, with different platelets content (5, 8, 12 and 20 vol%), have been obtained by controlled flocculation at a pH value in the range 6–7. From these powders, very close to theoretical density ($\geqslant$ 99% d_{th}) and slightly oriented platelet-reinforced alumina compacts have been obtained by hot pressing at temperatures ranging from 1500–1550 °C.

1. Introduction

Ceramics are increasingly considered to produce cutting tool tips, wear parts and structural components because of their thermomechanical properties, low density and excellent corrosion/oxidation resistance compared with hard metals and superalloys. However, the intrinsic brittleness of monolithic ceramics has limited their use in critical load-bearing structural components.

The reliability of ceramic materials can be improved by designing new materials with enhanced mechanical properties. In this way, whiskers [1–5], platelets [5–10] and powders [10, 11] are incorporated into the matrix to produce reinforced composites. Although the best mechanical behaviour is achieved when whiskers are employed, platelets are now increasingly used owing to the simplicity of handling, because they are not dangerous to health, as are whiskers [12], and because of the possibility of introducing higher contents without agglomeration problems.

The major objective of this investigation was to determine the optimum processing conditions which lead to homogeneous composites free of platelet agglomerates. In this way colloidal processing is considered as a potential method to improve particle/platelet dispersion. The suspension stability has been determined by measuring the zeta potential and viscosity versus pH, as well as the effect of deflocculants, solids content and platelet content on viscosity.

Because well-dispersed suspensions are important to obtain desirable green microstructures, segregation of platelets during consolidation must also be prevented to avoid inhomogeneities and reduced mechanical properties. In the present work, instantaneous flocculation has been used to achieve homogeneous powder/platelet mixtures.

From the powders obtained after drying the mixtures, platelet-reinforced alumina composites have been prepared using both pressureless sintering and hot pressing.

2. Experimental procedure

Al_2O_3 (T1, Atochem, France) and SiC (SF, C-Axis Technology, Canada) platelets were used as reinforcement phases. A submicronic alumina powder ($d_{50} \approx 0.3$ μm) was selected as the starting matrix material (CS-400, Lonza Martinswerk, Germany). Particle-size distributions were determined by X-ray sedimentation method using a Sedigraph 5000ET (Micromeritics, USA). The platelet length and thickness were also evaluated from the scanning electron micrographs by using an image analyzer (Morphomat 30, Zeiss, Germany). The specific surface area was determined by BET nitrogen gas adsorption (Monosorb MS-13, Quantachrome, USA). In Table I the chemical analysis and morphological characteristics of the starting materials are shown.

Zeta potential measurements as a function of suspension pH values were performed to determine the isoelectric point of the Al_2O_3 powder by using a mass transport analyzer (Micromeritics Inc, USA).

The rheological behaviour of the aqueous suspensions of alumina powder versus solids content, pH, deflocculants and platelets content was studied using a rotational viscosimeter (Haake Rotovisco RV 20, Germany). The alumina suspensions were prepared by mixing the powder with deionized water at 30 wt% solids loading. The pH was adjusted using HCl.

Agitation in an ultrasonic bath with a further treatment in an alumina ball mill for 2 h was selected in the present work to break up the agglomerates and to achieve optimum dispersion of the powder/platelet suspensions, Al_2O_3/Al_2O_3-pl and Al_2O_3/SiC-pl. The stable suspensions were suddenly flocculated by pH variation with HCl, then dried at 120 °C for 24 h and sieved up to 100 μm. The powders obtained by this procedure were: (i) hot-pressed in an argon atmosphere at 1500 °C for 0.5 h at 50 MPa applied pressure except the composite with 20 vol% SiC platelet which was hot pressed at 1550 °C for 1 h; (ii) isostatically pressed at 200 MPa and pressureless sin-

tered at 1650 °C for 1.5 h. Route (ii) was used to prepare Al_2O_3/Al_2O_3-pl composites.

Final densities were measured by Archimedes immersion technique in water.

The microstructure of the samples was analysed on polished surfaces by optical microscopy and on fracture surfaces by scanning electron microscopy.

3. Results and discussion

The particle electrophoresis results corresponding to Al_2O_3 powder are plotted in Fig. 1. As observed, the isoelectric point takes place at pH ~ 7.0.

The relative viscosity of 70 wt % alumina suspensions as a function of pH at a shear rate of 550 s^{-1}, is shown in Fig. 2. The maximum viscosity is observed at pH ~ 6.0. As expected, this pH value is in agreement with that of the isoelectric point (iep) (Fig. 1). A weight loss of ~ 1.0% at 500 °C was detected in the Al_2O_3 powder by thermogravimetric analysis (TG) (Fig. 3). This is due to the presence of organic processing additives which may be responsible for the low pH_{iep} obtained (Fig. 1). A maximum in zeta potential and a minimum in viscosity were observed at pH ~ 10. In the pH range 6–7, the suspensions flocculated due to the low zeta potentials, which is reflected in a high viscosity.

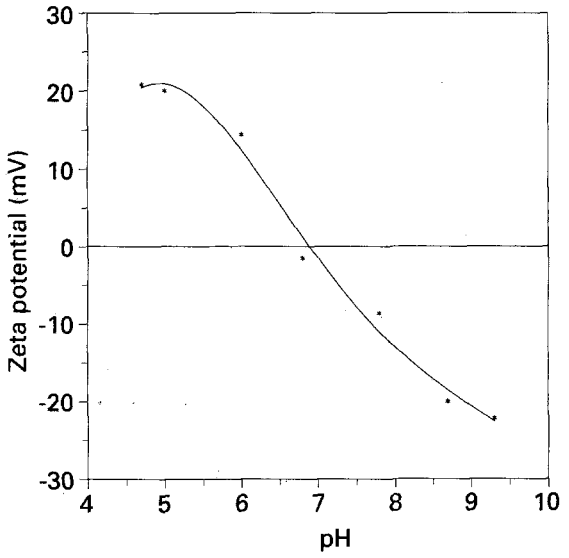

Figure 1 Zeta potential as a function of pH for Al_2O_3 powder.

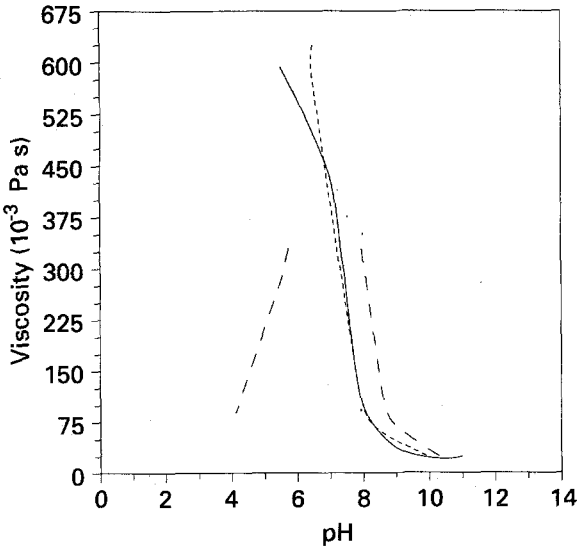

Figure 2 Relative viscosity of alumina suspensions (70 wt % solids content) versus pH at shear rate of 550 s^{-1} (——). 0 vol % platelets, (– – –) 5 vol % Sic platelets, (- - -) 5 vol % Al_2O_3 platelets.

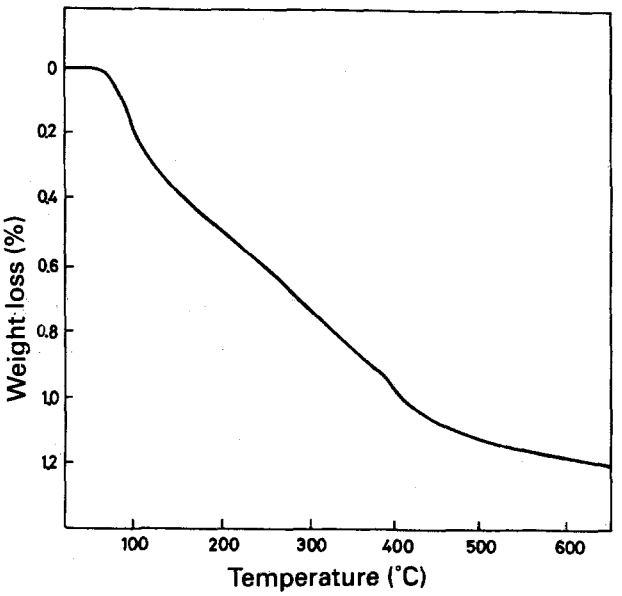

Figure 3 TG analysis of the starting Al_2O_3 powder.

Suspensions with high solids loading must be considered in order to minimize particle and platelet segregation. As shown in Fig. 4, the relative viscosity of Al_2O_3 suspensions increases with solids content. A 70 wt % solids loading was selected to prepare all the suspensions because of the good balance between low viscosity and high solids content.

The viscosity of alumina suspensions was also studied as a function of deflocculant content (Dolapix PC33, Schimmer-Schwarz, Germany). The results, plotted in Fig. 5, clearly show that this deflocculant does not improve the suspension viscosity behaviour.

The viscosity versus pH curves of Al_2O_3 powder, Al_2O_3/Al_2O_3 platelets and Al_2O_3/SiC platelets suspensions are shown in Fig. 2. It can be observed that the lowest viscosity is obtained at pH = 10 for all the suspensions.

The platelet content does not affect significantly the rheological behaviour of the alumina powder; the viscosity shows a slight decrease as platelet content increases (Fig. 6).

In summary, it can be stated that the optimal conditions to prepare highly dispersed suspension were found to be: (i) obtention of a stable suspension at pH ~ 10 with 70 wt % solids content, (ii) flocculation of the suspension by pH variation at pH ~ 7.

Although a well-controlled rheological behaviour is required to ensure uniform distribution of the platelets in the matrix, the agglomerates present in the starting platelets must be eliminated because they modify the microstructure and decrease the mechanical properties. Agitation in an ultrasonic bath and ball milling for 2 h have been proved to be adequate conditions to break down the possible platelet clusters and to

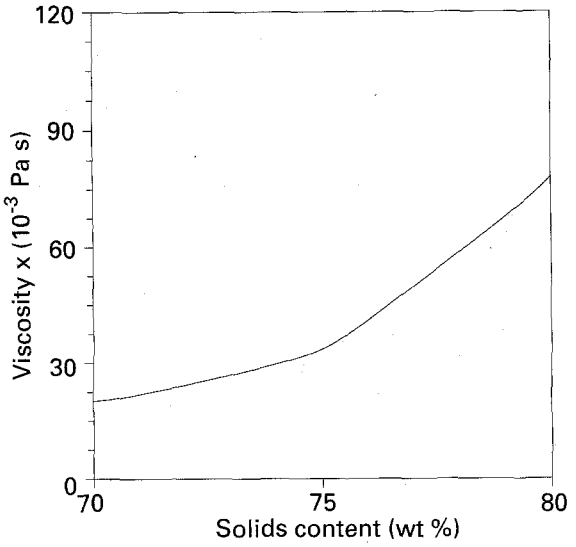

Figure 4 Relative viscosity of Al_2O_3 powder suspensions plotted against solids content at a shear rate of 550 s^{-1}.

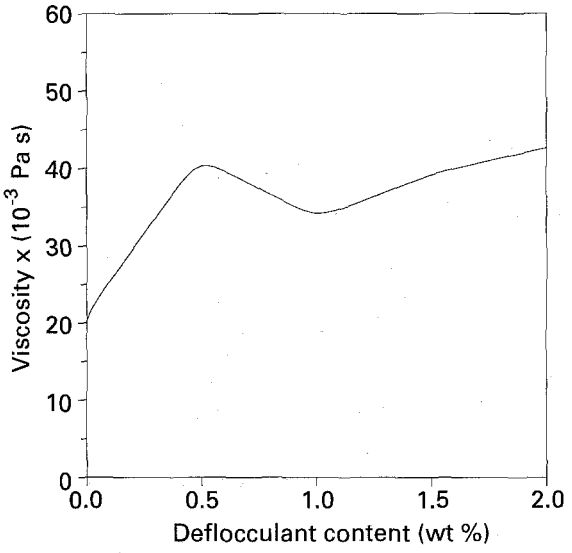

Figure 5 Relative viscosity of Al_2O_3 powder suspensions plotted against deflocculant content at a shear rate of 550 s^{-1}.

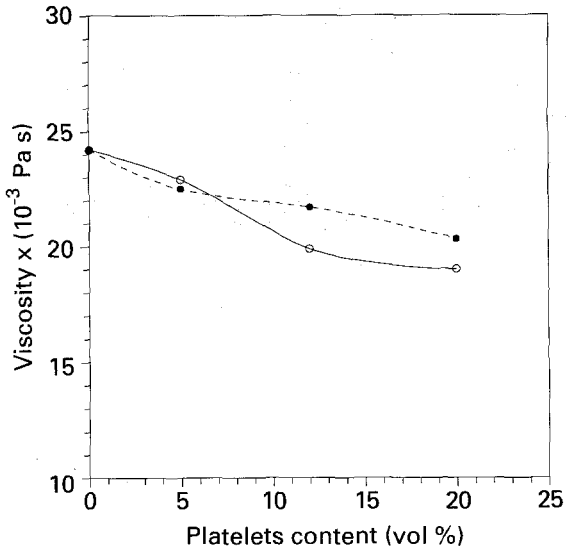

Figure 6 Relative viscosity of alumina/platelet ((●) Al_2O_3 or (○) SiC) suspensions as a function of platelets content at a shear rate of 550 s^{-1}.

achieve a good dispersion of the platelets. Fig. 7 shows the microstructure of Al_2O_3/Al_2O_3 platelet (5 vol %) composites obtained by pressureless sintering, starting from a mixture obtained according to the proposed homogenization treatment and from a mixture obtained by an alternative route in which only a high shear mixer is used for dispersion. It can be observed that with the selected method, no platelet clusters are detected and a more homogeneous microstructure is obtained.

The final densities of both SiC and Al_2O_3 platelet composites are summarized in Table II. The sintering behaviour of platelet-reinforced composites may be compared to that observed in bimodal distribution of spherical particles [13]. Fine particles show rapid densification because of their high curvature, while the large particles (platelets) exhibit a lower driving force for sintering. Consequently, pore channels between platelet and dense fine-grained regions appear, and then the relative density of the compacts decreases

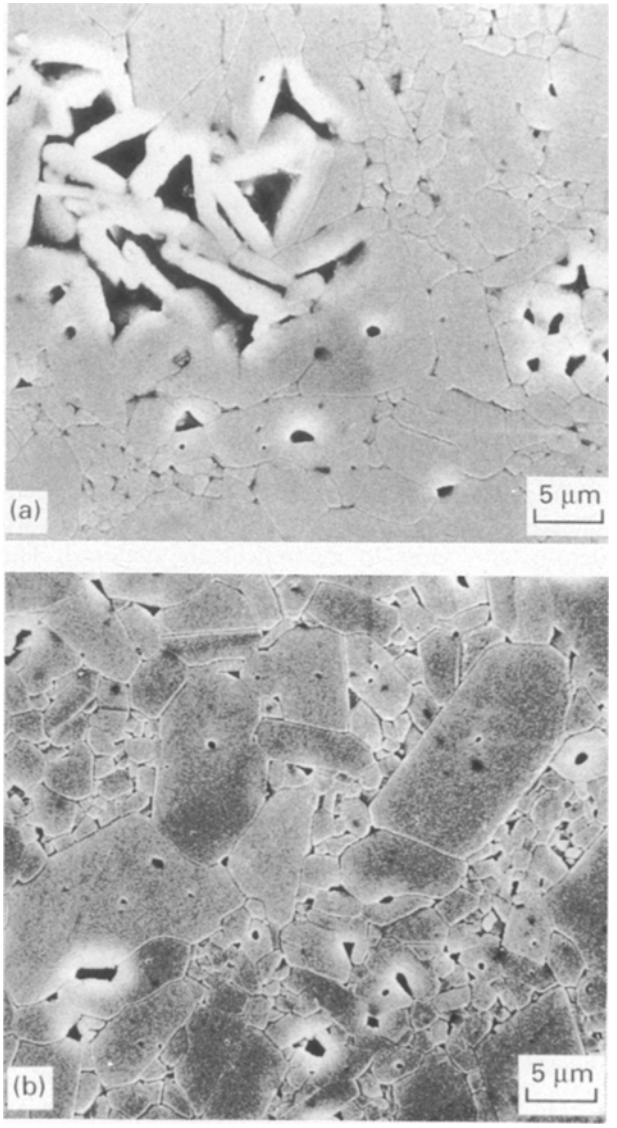

Figure 7 Scanning electron micrographs of Al_2O_3/5 vol % Al_2O_3 platelet composites sintered at 1600 °C for 1.5 h and thermally etched at 1400 °C/1.5 h obtained by: (a) mixing with a high shear mixer, and (b) agitation in an ultrasonic bath and ball milling for 2 h.

TABLE I Characteristics of the starting materials

	Al_2O_3 powder[a]	Al_2O_3 plat.	SiC plat.[a]
d_{50}[b] (μm)	~ 0.3	~ 4.3	~ 10.5
Diameter[c] (μm)	–	7.2	16.8
Thickness[c] (μm)	–	0.7	2.9
Aspect ratio	–	10.3	5.8
S_s(BET, m^2/g^{-1})	9	1.3	0.7
Chemical analysis (wt %)			
SiO_2	0.05	0.45	–
Al_2O_3	–	–	1.1
Fe_2O_3	0.03	0.08	0.014
TiO_2	0.005	0.04	0.003
CaO	0.05	0.02	0.01
MgO	–	0.007	0.003
NiO	–	–	0.001
LiF	–	0.72	–
Na_2O	0.1	0.32	–

[a] Reported impurities.
[b] Measured by Sedigraph.
[c] Measured by image analysis on scanning electron micrographs.

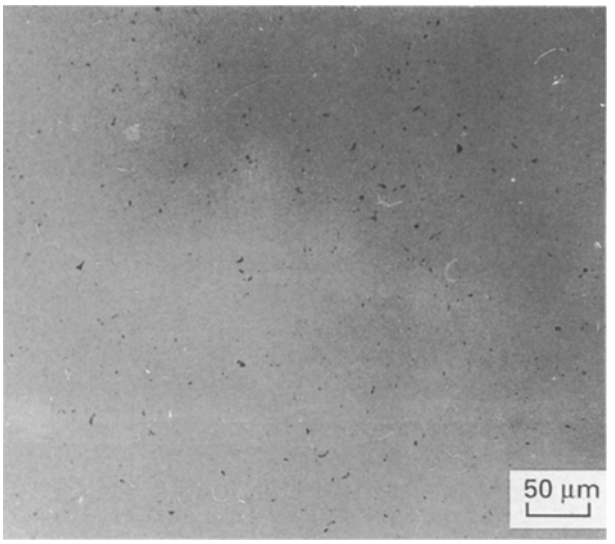

Figure 8 Optical micrograph of Al_2O_3/12 vol % Al_2O_3 platelet composite obtained by hot pressing at 1500 °C for 30 min and 50 MPa in argon.

TABLE II Relative densities of Al_2O_3/platelets composites

Platelets content (vol %)	% theoretical density, (d_{th})		
	Al_2O_3/Al_2O_3[a]	Al_2O_3/Al_2O_3[b]	Al_2O_3/SiC[b]
0	97.5	99.5	99.5
5	96.5	99.8	99.8
8	96.5	99.9	99.8
12	95.7	99.9	99.9
20	–	–	99.9

[a] Pressureless sintered at 1650 °C/1.5 h/air.
[b] Hot-pressed at 1500 °C/0.5 h and 1550 °C/1 h for platelet content of 0–12 and 20 vol %, respectively.

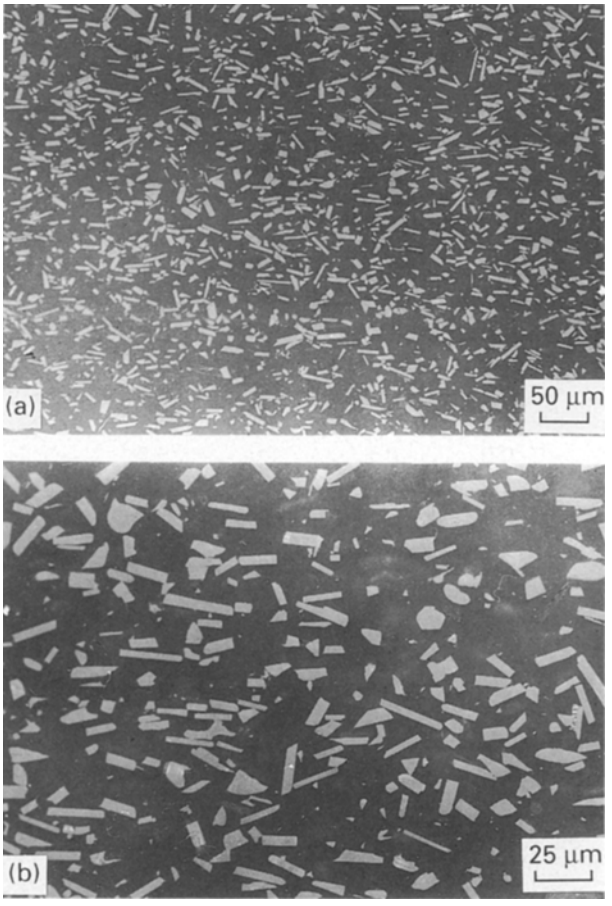

Figure 9 (a, b) Optical micrographs of Al_2O_3/20 vol % SiC platelet composites obtained by hot pressing at 1500 °C for 1 h and 50 MPa in argon.

when platelets are present. By hot pressing, the densification driving force increases and compacts with density very close to theoretical are obtained. The slightly lower density (99.5 wt %) obtained in the alumina compact compared with the densities obtained in both platelet-reinforced (Al_2O_3-pl and SiC-pl) compacts obtained by hot pressing (Table II) may be due to the presence of a small amount of glassy phase developed at high temperature as a consequence of the impurities introduced by both types of platelet (Table I).

Optical micrographs corresponding to Al_2O_3/Al_2O_3-pl and Al_2O_3/SiC-pl composites are shown in Figs 8 and 9. An excellent dispersion of the platelets in the alumina matrix and a microstructure completely free of pores can be observed. During hot pressing, platelets were slightly oriented with their diagonal essentially randomly oriented in the plane perpendicular to the hot-press direction (Fig. 9).

4. Conclusions

It has been proved that the colloidal route provides homogeneous platelet-reinforced composites free of agglomeration defects. Controlled flocculation opens the possibility of maintaining a well-dispersed structure without segregation by suddenly increasing the viscosity of the slip. Agitation in an ultrasonic bath and alumina ball milling are required to break down the platelet clusters.

Relative densities of 97% d_{th} have been obtained by pressureless sintering for Al_2O_3/Al_2O_3-pl composites. The use of hot pressing allows relative densities very close to the theoretical value (> 99.5%) to be achieved for both Al_2O_3 and SiC platelets.

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