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The processing of monodisperse boron-doped SiO_2 particles*

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Two processing methods were developed to add B_2O_3 to monodispersed, spherical (0.4 μm in diameter) SiO_2 particles. In each case the average bulk concentration of 1.5 wt% B_2O_3 was added to the surface of the silica particles. In the first process B_2O_3 was added with a boron alkoxide, $B(OC_4H_9)_3$, during the precipitation of monodispersed silica particles; in the second process boric acid was precipitated on the surface of monodispersed silica particles from aqueous solutions. The particles were settled, dried, and then sintered at temperatures from 1000 to 1100° C. Several problems were encountered in these steps, but it was possible to sinter thin pieces ~500 μm thick of boron-doped silica particles at 1000° C in 1 h. Also, cakes ~20 to 30 mm thick of boron-doped particles coated with boric acid and a sodium salt (to add Na_2O) sintered to nearly full density at 1050° C in less than 2 h. It was not possible to form sintered, continuous thin films ~1 to 5 μm thick of boron-doped silica from casts of particles on a substrate, due to shrinkage cracks.

1. Introduction

Maintaining reproducibility and reliability has been a major problem in the production of high-technology ceramics, especially in the electronics industry. Much research has been devoted to gaining more complete control over the sintering process. It is now understood, however, that even the best sintering techniques cannot overcome problems inherent to the green microstructure such as low green density, agglomeration, and major voids. Therefore, research efforts have begun to focus on the processing of ceramic particles to form more "ideal" powders and more uniform and reproducible green microstructures.

The ideal starting material to fabricate a reproducible, fine-grained ceramic is a small-sized (0.1 to 1.0 μm), equiaxed powder with a narrow size distribution and acceptable handling characteristics which can be fabricated into dense, uniform green bodies. Colloid chemists have developed techniques to form a number of oxide and hydroxide single-component particles with these characteristics by precipitation in dilute aqueous systems [1–10]. The next step in these research efforts is to develop methods for adding a second component to one of these aqueous systems.

It has been shown [1, 11] that monodispersed, spherical SiO_2 particles 0.1 to 1.0 μm in diameter can be made easily by the hydrolysis of silicon alkoxides. The purpose of this research was to study the addition of a second component to the SiO_2 particles while maintaining ideal powder properties. The B_2O_3–SiO_2 system, which has potential use in the electronics industry as an insulation-layer material, was chosen for study. The goal of this research, then, was to develop processing methods to add B_2O_3 to ideal SiO_2 particles and to study the effects on sintering of a monodispersed B_2O_3–SiO_2 powder. A further goal was to study the casting and sintering of 1 to 5 μm films of boron-doped silica particles.

2. Experimental procedure

2.1. Formation of silica particles

Stöber *et al.* [1] and Huynh [11] studied the formation of SiO_2 particles by the hydrolysis of silicon alkoxides. From these studies a specific system was chosen which produced very monodispersed, spherical SiO_2 particles ~0.4 μm in diameter.

The particles were formed by mixing specific amounts of ethanol and NH_4OH at room temperature in a 250 ml Pyrex crystallizing dish. This mixture was continuously stirred using a magnetic stirrer (Magnestir, Lab-Line Instruments Inc., Melrose Park, Illinois) with a 1.5 inch stirring bar rotating at approximately 60 revs min^{-1}. The silicon alkoxide, tetraethoxysilane (TEOS, $Si(OC_2H_5)_4$), was then added all at once to this mixture while agitation was continued. All the alkoxides, the ethanol, and the ammonium hydroxide were filtered through a 0.4 μm Type BD Millipore (Millipore Corp., Bedford, Massachusetts) filter before use. All glassware was cleaned with 1 M hydrofluoric acid and rinsed with doubly deionized water.

After the TEOS was added the dish was tightly covered with foil to avoid any evaporation losses. The beginning of the hydrolysis reaction was marked by opalescence of the solution. As the particles grew the solution became more turbid. Agitation was continued

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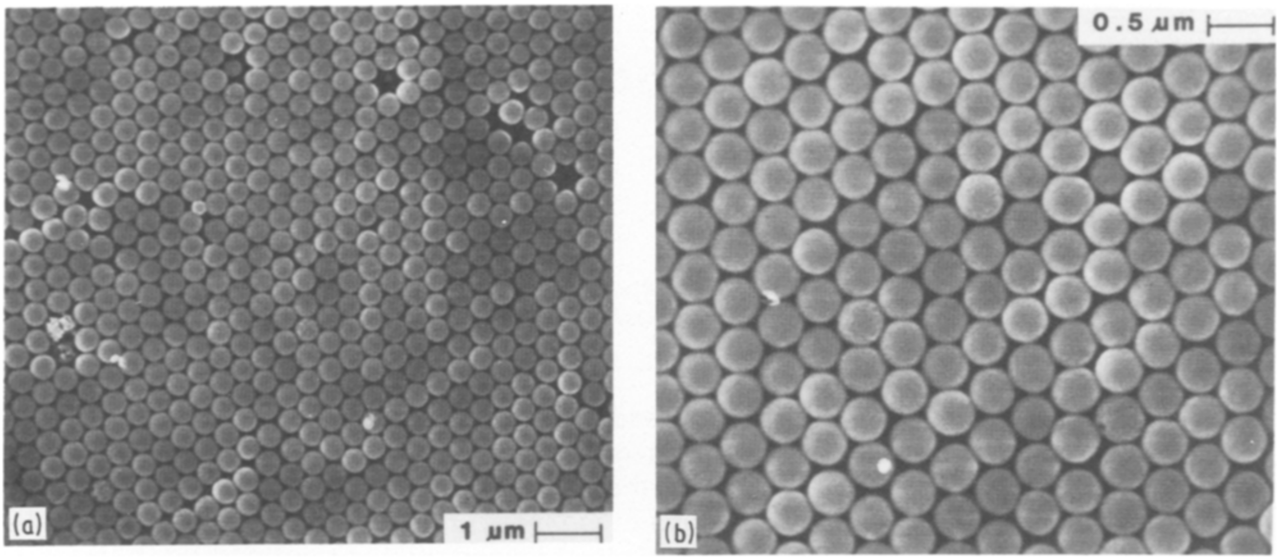

Figure 1 Scanning electron micrographs of SiO_2 particles at two magnifications.

for 24 h to ensure full particle growth. The specific formulation used in these studies was 1.95 M NH_3, 0.30 M TEOS, and 4.52 M H_2O in the final solution. The standard formulation in volume quantities was 90 ml ethanol, 14.25 ml NH_4OH, and 7.5 ml $Si(OC_2H_5)_4$. It was not necessary to add any extra water to the ammonium hydroxide. For this particular formulation, the initial reaction occurred about 4 min after the addition of the TEOS. The particles grew to 91% of their final diameter after 1 h.

After 24 h of stirring, the solution was centrifuged in 50 ml polyethylene centrifuge tubes at ~2000 g (3000 r.p.m). The NH_4OH/ethanol mixture was then decanted and discarded when centrifugation was complete. The particles in the centrifuge tube were redispersed in water or ethanol by ultrasonication (Sonicator Model W-22F, Ultrasonics Inc., Long Island, New York). They were stored in this form until used. Fig. 1 shows scanning electron micrographs (SEM) of the SiO_2 particles produced by this method.

2.2. Boron addition

Boron was added to the silica particles by two methods: in the first, a boron alkoxide was incorporated into the hydrolysis process for forming SiO_2 particles; in the second, boric acid was precipitated on the surface of silica particles from an aqueous solution.

2.2.1. Boron alkoxides in the hydrolysis process

After some experimenting a method was devised to add boron to silica particles throughout the hydrolysis process. First, the standard silica formulation given below was used to make the silica particles. After 0.5 h of growth ($d = 0.34\,\mu m$), an ethanol–tri-n-butyl borate ($(B(OC_4H_9)_3)$) solution was slowly dripped into the reaction medium while stirring continuously. The dripping process took approximately 0.5 h to complete. As before, stirring was continued for 24 h. The standard formulation for this process (in the final solution) was as follows: 0.27 M $Si(OR)_4$, 1.76 M NH_3, 0.52 M $B(OC_4H_9)_3$, and the balance ethanol.

The $B(OC_4H_9)_3$ was diluted with ethanol 1 : 5.75, then was added slowly to the reaction medium. The standard formulation in volume quantities was:

SiO_2: 90 ml ethanol, 14.25 ml NH_4OH, 7.5 ml $Si(OC_2H_5)_4$

Boron solution: 1.74 ml $B(OC_4H_9)_3$, 10 ml ethanol

2.2.2. Boric acid solutions

Aqueous solutions of orthoboric acid, H_3BO_3, were also used to add boron to silica particles. When the boric acid solution was added to a settled cake of SiO_2 particles, the interstices of the cake were filled with the solution. When the cake was dried, the water evaporated and the boric acid precipitated on the surfaces of the particles. When heated further the boric acid dehydrated to form B_2O_3. The solubility of boric acid in water at room temperature is 6.35 g/100 ml H_2O (1.03 M). One molar solutions were used for these experiments. Because one molar solutions are not very concentrated, only a small amount of B_2O_3 can be added by this method. Therefore this procedure was used to supplement the B_2O_3 added in the hydrolysis process.

2.3. Sodium chloride solutions

Sodium chloride solutions were also added to sediment cakes in the same manner as the boric acid solutions. They were added for two reasons. First, they are a source of sodium ions, which are an additional sintering aid because they lower the viscosity of SiO_2 [12], as does B_2O_3 [13]. Second, a salt solution added to a sediment cake will help to increase the green density and strength by collapsing the electric double layers of the particles and allowing the particles to come in closer contact. The concentration of the NaCl solutions was 3.00 M; they were added in conjunction with the boric acid solutions.

2.4. Settling and drying

The particle dispersions were settled to form cakes or films. In most of the initial experiments settling and drying were carried out in centrifuge tubes. But

TABLE I Boron alkoxides in the hydrolysis process.

Experiment number	Formulation (in volume quantities)	Method of addition	Result
Standard silica formulation (SSF)	90 ml ethanol 14.25 ml NH_4OH 7.5 ml $Si(OC_2H_5)_4$ (TEOS)		Monodisperse 0.4 μm particles
1	SSF + 0.5 ml $B(OC_2H_5)_3$	TEOS + $B(OC_2H_5)_3$ mixed together and added all at once	Not monodisperse
2	SSF + 0.5 ml $B(OC_2H_5)_3$	$B(OC_2H_5)_3$ added after 5 min of silica particle growth	White fluffy precipitate formed upon addition of $B(OC_2H_5)_3$
4	$SSF + \begin{cases} 0.5\,\text{ml } B(OC_2H_5)_3 \\ 12.5\,\text{ml ethanol} \end{cases}$	$B(OC_2H_5)_3$–ethanol mixture added dropwise slowly after 5 min of silica particle growth	Not monodisperse
8	SSF + 1.4 ml $B(OC_3H_7)_3$	$B(OC_3H_7)_3$ added after 0.5 h of silica particle growth	B_2O_3 particles hydrolysed separately
9	SSF + 1.74 ml $B(OC_4H_9)_3$	$B(OC_4H_9)_3$ added after 0.5 h of silica particle growth	B_2O_3 particles hydrolysed separately
10	$SSF + \begin{cases} 1.40\,\text{ml } B(OC_3H_7)_3 \\ 10\,\text{ml ethanol} \end{cases}$	$B(OC_3H_7)_3$–ethanol mixture added dropwise slowly after 0.5 h of silica particle growth	Monodisperse 0.4 μm particles 1.5 w/o B_2O_3
11	$SSF + \begin{cases} 1.74\,\text{ml } B(OC_4H_9)_3 \\ 10\,\text{ml ethanol} \end{cases}$	$B(OC_4H_9)_3$–ethanol mixture added dropwise slowly after 0.5 h silica particle growth	Monodisperse 0.4 μm particles 1.6 w/o B_2O_3

because the centrifuge cakes tended to crack excessively during sintering, later experiments used flat-settled cakes, which did not crack as much.

Settling to form thick cakes ~20 to 30 mm thick was done in Nalgene bottles in which the bottoms were cut out and replaced with Parafilm (American Can Co., Greenwich, Connecticut) or Teflon (Berghof/America Inc., Raymond, New Hampshire) film. The films were held in place by rubber stoppers. This allowed easy removal of the resulting cake. After the particles had settled, any salt or boric acid solutions were added to the supernatant. The boric acid and salt solutions were allowed to sit with the sediment for 1 h. After that, as much of the supernatant as possible was siphoned off. The cakes were dried in air until they could be removed from the containers, then at 70° C in air for 24 h, and finally in vacuum (< 1 torr) for an additional 10 h. Thin cakes were settled and dried in plastic petri dishes.

In addition, thin films 1 to 5 μm thick on fused quartz were used for some sintering studies. These films were formed by dripping a concentrated ethanol/particle dispersion onto the substrate and drying as described above. Ethanol dispersions were used in this case because of the wetting characteristics of ethanol on fused quartz. Water dispersions did not wet well and tended to bead up, while ethanol dispersions wet the quartz fairly well and allowed a relatively even film of particles to form.

2.5. Sintering

In all cases sintering was carried out in air in a box furnace (Blue M. Electric Co., Blue Island, Illinois) heated by Kanthal windings. Sintering temperatures were measured with a chromel–alumel thermocouple and maintained within ±5° C of the set point. All of the samples were sintered at temperatures between 1000 and 1100° C. Samples were cooled in the furnace to avoid thermal shock. Fused quartz and platinum were used as substrates.

2.6. Characterization techniques

The following techniques were used to characterize the particles:

1. Chemical analysis: The boron content of the particles was measured with an Ebert 3.4 m Emission Spectrograph (Jarrel-Ash Co., Waltham, Massachusetts). Five samples were analysed and the average value was reported. The variation in readings was ±0.2 wt % B_2O_3.
2. Density: The density of the powders was measured with a stereopycnometer (Quantachrome Stereopycnometer, Quantachrome Corp., Syosset, New York) using helium gas. Four measurements were made on each sample. The variation in readings was less than 0.5%.
3. Surface area: Single-point BET analysis (Quantachrome Quantasorb, Quantachrome Corp., Syosset, New York) with nitrogen gas was used to measure the surface area of the particles. All powders were outgassed in flowing nitrogen gas at 200° C for 16 h. Five readings were recorded for each sample and averaged. The variation in readings was less than 1%.

TABLE II Particle properties.

Particles	B_2O_3 (wt %)	Density ($g\,cm^{-3}$)	Real particle diameter (μm)	BET surface area ($m^2\,g^{-1}$)
SiO_2	0	2.04	0.41	11.0
B_2O_3–SiO_2	1.6	2.01	0.40	10.0

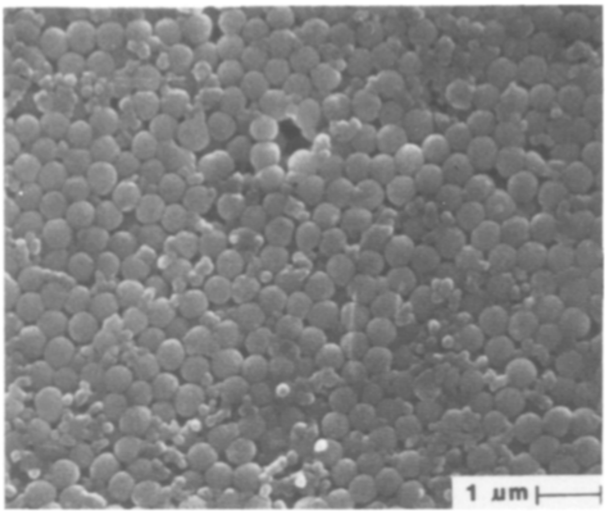

Figure 2 Scanning electron micrograph of the top surface of a centrifuge cake of particles made in Experiment 9.

4. Scanning electron microscopy: For observation in the SEM the samples were coated (Hummer II Evaporator, Technics, Alexandria, Virginia) with 20 nm of Au–Pd. A JEOL JSM-35 (Japanese Electron Optics Laboratory Tokyo) SEM was used to view the samples.

3. Results and discussion

Two methods were developed to add B_2O_3 to monodispersed SiO_2 particles. This section will discuss the two methods and the characteristics of boron-doped particles.

3.1. Boron addition in the hydrolysis process

Table I lists the experiments in which boron alkoxides were added to silica particles during hydrolysis. The important variables in determining the success or failure of these trials were time of addition, chain length, and concentration of the boron alkoxide.

3.1.1. Time of addition

Because B_2O_3 additions to SiO_2 decrease viscosity and thereby improve sinterability, it is desirable to have the B_2O_3 concentrated at the surface of the particles. Also, Experiments 1 and 4 indicated that if the boron alkoxide is added too early in the process, the particles will not be monodispersed. Therefore, the time of addition of the boron alkoxide was arbitrarily chosen to be after 0.5 h of silica growth for most experiments. By this time about 84% of the final particle diameter has been achieved.

3.1.2. Chain length

Experiments 10 and 11 showed that it was necessary to add a long-chain alkoxide (butyl borate was chosen) to the silica particles to get monodispersed borosilicate particles. The short-chain borate $B(OC_2H_5)_3$ used in Experiments 1 to 9 hydrolysed quite rapidly forming separate B_2O_3 particles rather than co-hydrolysing with the silica particles. Experiment 2 was the best example of this: the $B(OC_2H_5)_3$ formed a fluffy precipitate of B_2O_3 when added to solution. The long-chain alkoxides hydrolysed more slowly, allowing the borosilicate particles to grow without separate hydrolysis.

3.1.3. Concentration

As can be seen from the formulations of Experiments 10 and 11 in Table I, it was necessary to dilute the boron alkoxide with ethanol before addition to the reaction vessel. Such a dilution serves two purposes. First, it allows slow, dropwise addition of the boron alkoxide, and second, it decreases the concentration of $B(OC_4H_9)_3$ to a level where it will not hydrolyse too rapidly. If the alkoxide is not dilute, as in Experiments 8 and 9, separate B_2O_3 particles precipitate out. These precipitates can be seen at the top of the centrifuge cake in Fig. 2.

3.2. B_2O_3–SiO_2 particles

As mentioned earlier, a standard formulation was chosen to form the borosilicate powders (Experiment 11 in Table I). Table II lists the properties of these particles and the SiO_2 particles. Scanning electron micrographs of the SiO_2 particles and the boron-doped SiO_2 particles are shown in Figs 1 and 3, respectively. The particles are small (~0.4 µm in diameter), spherical, and monodisperse.

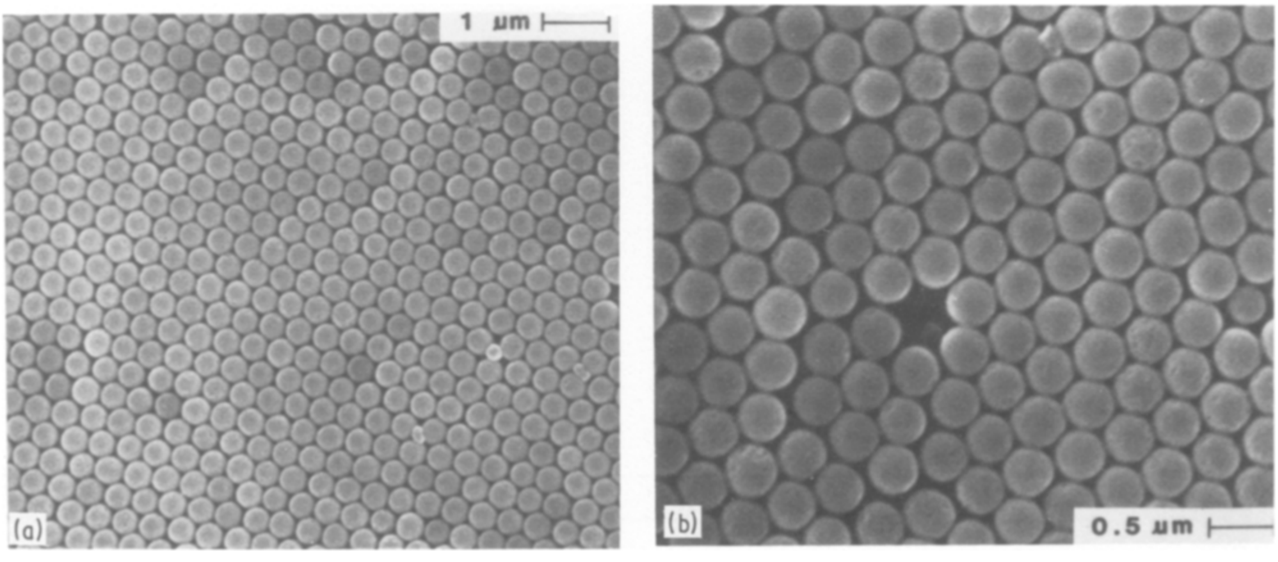

Figure 3 Scanning electron micrographs of boron-doped SiO_2 particles at two magnifications.

TABLE III The effects of increasing the boron content in boron-doped SiO_2

Experiment number	Formulation (in volume quantities)	Concentration	Result
Standard silica formulation (SSF)	90 ml ethanol 14.25 ml NH_4OH 7.5 ml $Si(OC_2H_5)_4$		Monodisperse 0.4 μm particles
11	$\text{SSF} + \left\{\begin{array}{l}\text{10 ml ethanol}\\ \text{1.74 ml } B(OC_4H_9)_3\end{array}\right.$		Monodisperse 0.4 μm particles 1.6 w/o B_2O_3
11A	$\text{SSF} + \left\{\begin{array}{l}\text{20 ml ethanol}\\ \text{3.5 ml } B(OC_4H_9)_3\end{array}\right.$	$\left(\frac{M_B}{M_S}\right)_A = 2\left(\frac{M_B}{M_S}\right)_{11}$	Coagulation upon addition of $B(OC_4H_9)_3$. Particles monodispersed, but coagulated
11B	$\text{SSF} + \left\{\begin{array}{l}\text{1.53 ml } NH_4OH\\ \text{20 ml ethanol}\\ \text{3.5 ml } B(OC_4H_9)_3\end{array}\right.$	$\left(\frac{M_B}{M_S}\right)_B = 2\left(\frac{M_B}{M_S}\right)_{11}$ $(M_N)_B = (M_N)_{11}$	Coagulation upon addition of $B(OC_4H_9)_3$. Particles not monodispersed
11C	$\text{SSF} + \left\{\begin{array}{l}\text{14.25 ml } NH_4OH\\ \text{20 ml ethanol}\\ \text{3.5 ml } B(OC_4H_9)_3\end{array}\right.$	$\left(\frac{M_B}{M_S}\right)_C = 2\left(\frac{M_B}{M_S}\right)_{11}$ $\left(\frac{M_N}{M_B}\right)_C = \left(\frac{M_N}{M_B}\right)_{11}$	Coagulation upon addition of $B(OC_4H_9)_3$. Particles monodispersed, but coagulated
11D	$\text{SSF} + \left\{\begin{array}{l}\text{122 ml ethanol}\\ \text{3.5 ml } B(OC_4H_9)_3\end{array}\right.$	$\left(\frac{M_B}{M_S}\right)_D = 2\left(\frac{M_B}{M_S}\right)_{11}$ $(M_B)_D = (M_B)_{11}$	Monodisperse 0.4 μm particles, no coagulation 2.45 w/o B_2O_3

Procedure: Ethanol–$B(OC_4H_9)_3$ mixtures added dropwise slowly after 0.5 h of silica particle growth. Any excess NH_4OH added immediately before the ethanol–$B(OC_4H_9)_3$ addition.

M_B = molarity of $B(OC_4H_9)_3$
M_S = molarity of $Si(OC_2H_5)_4$
M_N = molarity of NH_4OH

e.g. $\underset{\text{molarity}}{\left(\frac{M_B}{M_S}\right)_A} \overset{\text{trial}}{=} 2\left(\frac{M_B}{M_S}\right)_{11}$

The average boron content of the borosilicate particles was determined by emission spectroscopy to be 1.6 ± 0.2 wt % B_2O_3. Because the silica particles were allowed to grow for 0.5 h before the slow boron addition, the B_2O_3 should be concentrated in the outer 40% of the volume of particles.

Table III lists the experiments in which it was attempted to increase the boron concentration in the particles. Experiment 11D resulted in an increase in the average B_2O_3 concentration to 2.45 wt %. An unfortunate consequence, though, is that the yield is greatly reduced. In order to maintain a constant concentration of $B(OC_4H_9)_3$ while doubling the molar ratio of $B(OC_4H_9)_3$ to $Si(OC_2H_5)_4$, the amount of ethanol must be increased by 133%. Any further increases in the above ratio would be impractical because of this limitation.

3.3. Settling and drying

Fig. 4 is an SEM showing the well-ordered structures obtained by centrifuging the particles in water and then drying.

3.4. Sintering

One of the primary advantages of monodispersed particles is their ability to pack in a uniform, ordered,

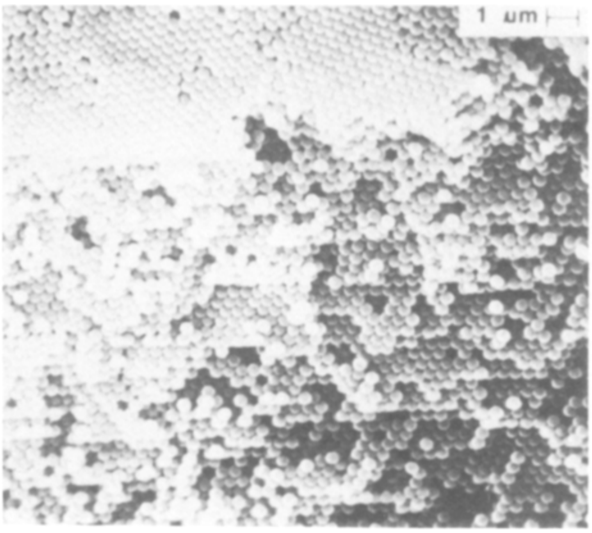

Figure 4 Scanning electron micrograph of a fracture surface of a centrifuge cake of borosilicate particles.

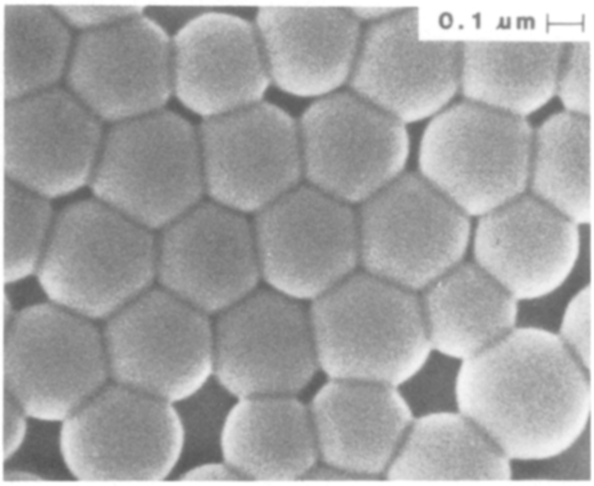

Figure 5 Scanning electron micrograph of partially sintered borosilicate particles with 120° dihedral angles.

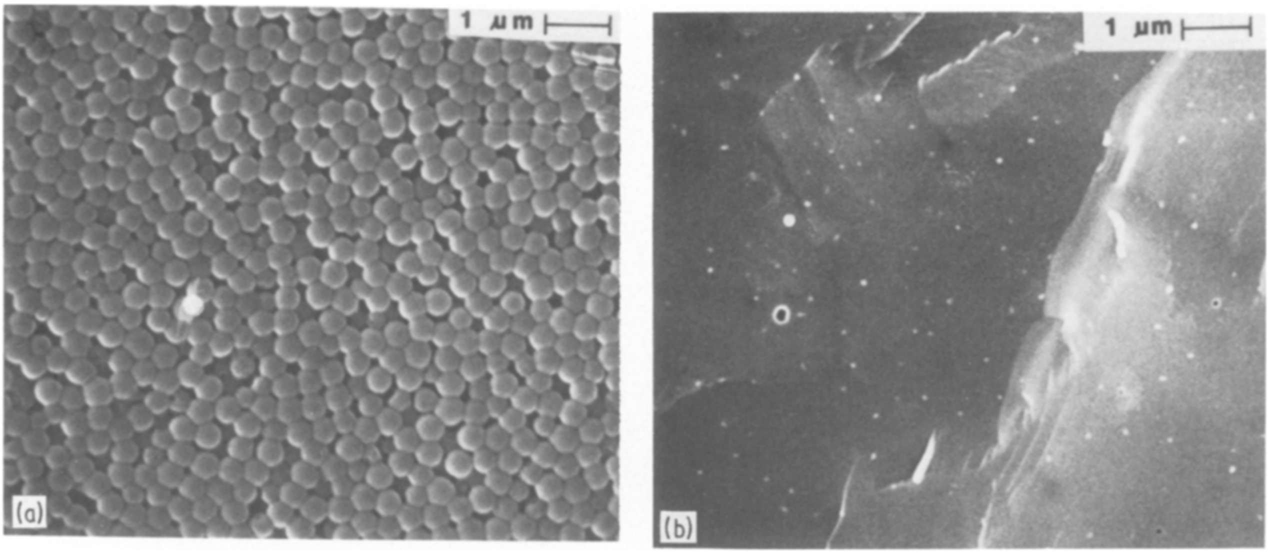

Figure 6 Scanning electron micrographs of a sintered thin cake of borosilicate particles: (a) top surface, and (b) fracture surface.

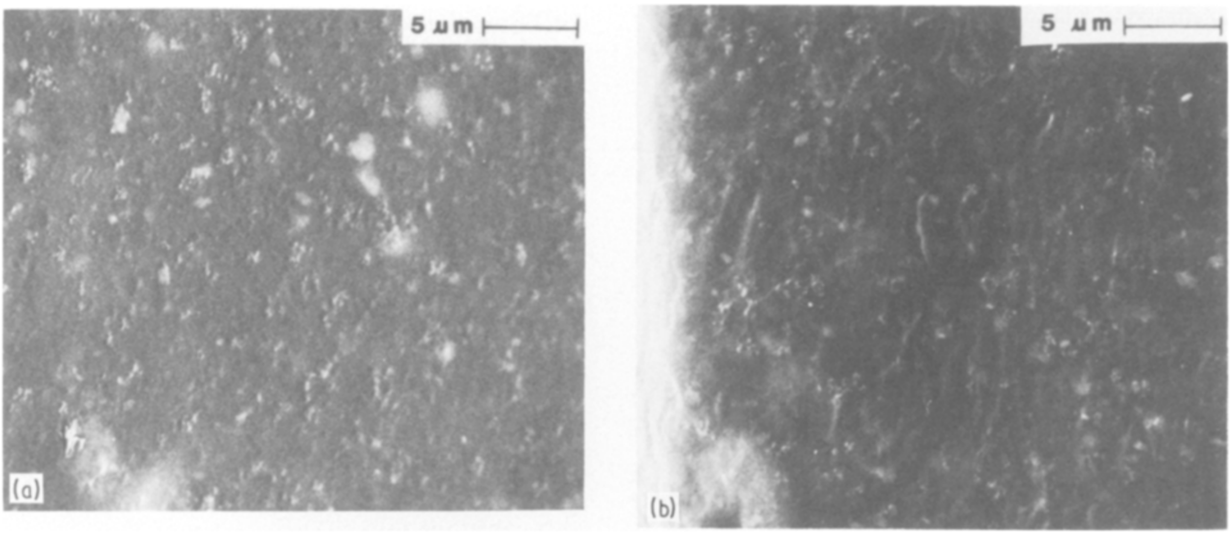

Figure 7 Scanning electron micrographs of a thin cake of borosilicate particles sintered at 1000° C for 1 h: (a) top surface, and (b) fracture surface.

and dense manner. This, of course, leads to better sintering control. In the development of sintering theory ideal particle packing must often be assumed, even though in most ceramic bodies this is not the case. With monodisperse powders, though, there is often a good portion of packing that is very near ideal. This results in sintered microstructures in which the ideal 120° dihedral angle is present. Fig. 5 shows a partially sintered microstructure of borosilicate particles with quite a few 120° dihedral angles. As the particles sinter further, the microstructure blurs. And because these are amorphous particles, the fully sintered microstructure will be completely smooth with no grain boundaries.

3.4.1. Sintering of cast borosilicate powders

In sintering studies on casts of the borosilicate powders a number of problems were evident. First, there was a high tendency for the samples to crack during sintering. Most of these cracks, though, could be traced to minute cracks that formed during the drying process. Second, thin pieces ~ 500 µm thick (cast in plastic petri dishes) sintered much better than thick pieces ~ 20 to 30 mm thick (cast in Nalgene containers). This may indicate differences in packing densities of the different pieces, which might result from variations in the containers and/or slurry concentration. Also,

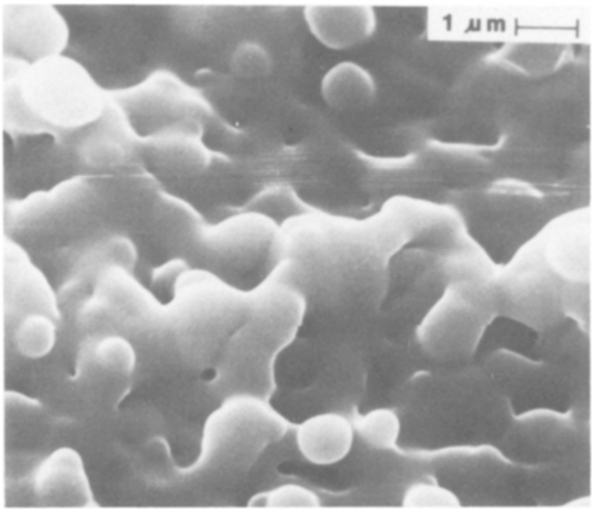

Figure 8 Scanning electron micrograph of loose borosilicate powder sintered at 1000° C for 2 h.

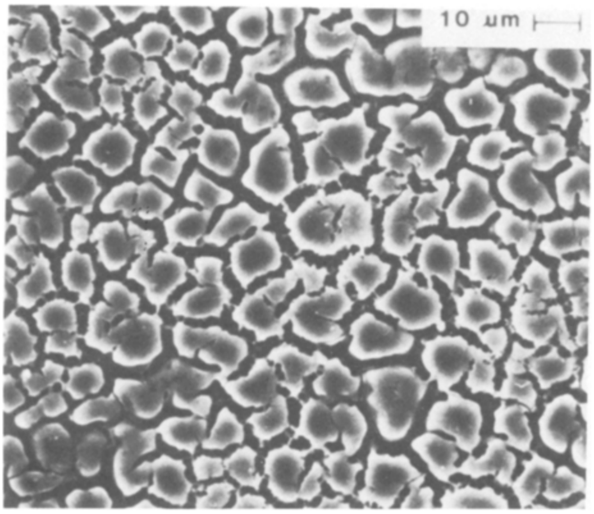

Figure 9 Scanning electron micrograph of a borosilicate film 5 μm thick sintered at 1000° C for 5 h.

because thin pieces have more exposed surface/unit mass than thick pieces, contaminations from the furnace walls will more strongly affect thin pieces. Another explanation could be gas transport effects, i.e., gas must be diffused away from the particle surface before sintering can begin. The removal of hydroxyl and alkyl groups from the surface of the particles might slow down the sintering of thick pieces. More study is needed to determine the cause(s) of this problem.

Finally, a third problem is illustrated in Fig. 6. In this case the top surface of a thin piece is barely sintered (Fig. 6a), while the fracture surface shows nearly complete sintering throughout the interior of the piece (Fig. 6b). The reason for this is not known although the atmosphere in the furnace may be responsible.

Despite these problems, thin pieces sintered to nearly full density in as little as 1 h at 1000° C. Fig. 7 shows the top and fracture surfaces of a piece sintered at 1000° C for 1 h. The piece is clearly well-sintered and dense. Fig. 8 shows some loose powder sintered at 1000° C for 2 h. Even in only 2 h these particles are well-sintered.

3.4.2. Sintering of films

The sintering of films 1 to 5 μm thick on fused quartz substrates was also studied. It was hoped that shrinkage could be controlled to only occur in the z-direction (one-dimensional shrinkage perpendicular to the substrate), while maintaining a continuous film in the $x-y$ direction. This was not possible, however, as can be seen in Fig. 9. Each small area was well-sintered, but there were large cracks between the areas. The $x-y$ shrinkage (estimated from the micrograph) was ~ 40%. Using relationships derived by Coble [14], it was calculated that a pressure of at least 430 p.s.i. (3 MPa) would be required to force shrinkage in the z-direction only.

3.4.3. Sintering with boric acid and salt

The effect on sintering of adding boric acid and salt to the boron-doped silica particles was very positive. Even thick pieces sintered to nearly full density in short periods of time. Fig. 10 shows micrographs of the top and fracture surfaces of a thick piece sintered for 2 h at 1050° C. Some of the texture on the top surface is a remnant from dripping the boric acid/salt solution onto the sediment cake. None of the problems mentioned in the section on sintering cast borosilicate powders were noticed in these samples. Apparently, in high enough concentrations, the additives strongly promote sintering.

Acknowledgements

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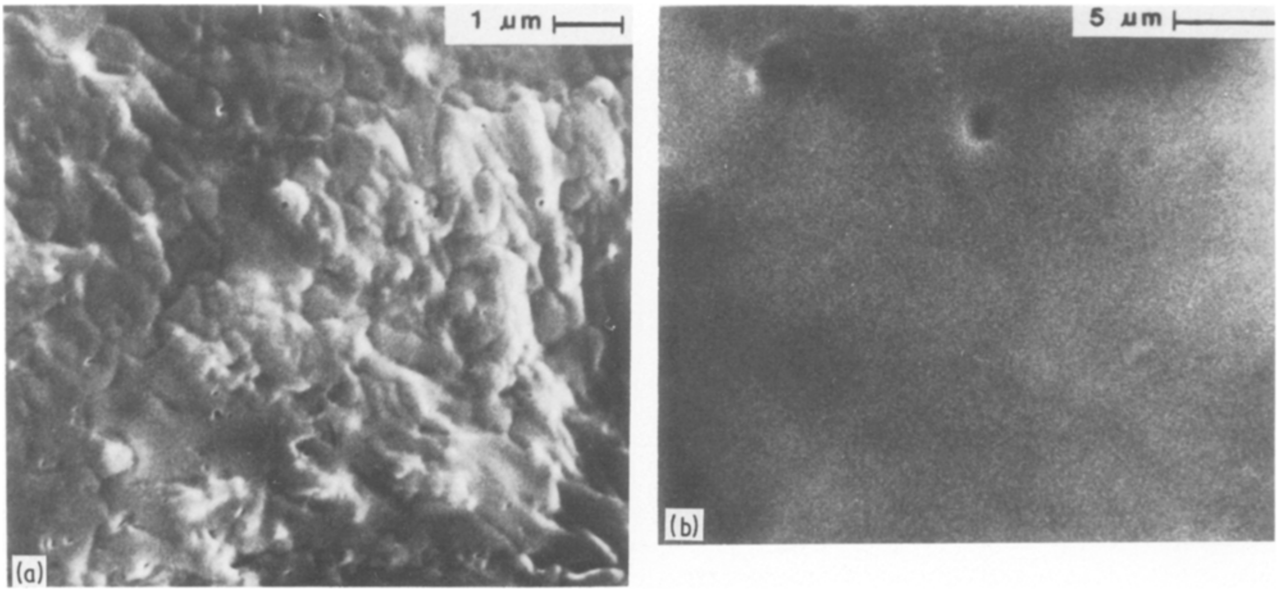

Figure 10 Scanning electron micrographs of a cake of borosilicate particles coated with boric acid and NaCl and sintered at 1050° C for 2 h: (a) top surface, and (b) fracture surface.

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