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Compaction Behaviour of Spray-Dried Silicon Carbide Powders

Dean-Mo Liu & Chen-Tsu Fu

Materials Research Laboratories, Industrial Technology Research Institute, Chutung, Hsinchu 31015, Taiwan

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Abstract: Compaction behaviour and resultant green density of spray-dried silicon carbide powder were investigated using a uniaxial die-pressing technique with compaction pressure ranging from 6·8 MPa to 154 MPa. Experimental results show that the green density of the silicon carbide powder compacts behaves as a function of organic phase content, temperature, relative humidity, powder size and its distribution; the density increases with relative humidity and with organic phase content. For a given quantity of organic phases, increase in temperature reduces the compact density when the compaction pressure is relatively low. For powders of approximately the same powder size distribution, increase in the mean powder size decreases the compact density at lower pressure regions; however, a higher density is obtainable at higher compaction pressures. The elastic recovery of the compacts after removal of the pressure was recorded, which became appreciable at pressure > ~100 MPa at temperatures below the glass transition temperature (T_g) of the organic phases.

INTRODUCTION

Consolidation of ceramic powders by means of die-pressing is the most widely used method for industrial production. This method usually requires ceramic powders with high flowability to facilitate the filling of the die cavity. This can be achieved by using a conventional spray-drying technique which produces powders with near-spherical morphology and greater than ~20 μm in size. Sometimes, the spray-dried powders may contain desired organic phases such as binder, plasticizer, and sometimes lubricant, for specific purposes. The investigation of the compaction behaviour of ceramic powders is not extensive,[1-6] probably because of its simplicity in forming technique. However, in spite of the factors such as organic phase content, temperature, powder size, and relative humidity, etc., which affect the compaction behaviour of the powders, the compaction response diagram (i.e. relative density versus logarithm of pressure) for most powder granules can generally be described by a model proposed by Lukasiewicz and Reed,[7] as schematically illustrated in Fig. 1. At relatively low pressure regions (stage I), the density changes slightly with increasing pressure, up to a breakpoint pressure (i.e. powder yield pressure) at which the density–compaction pressure slope increases (stage II) with a value depending upon powder characteristics and environmental conditions. In stage II, the increase of compact density is due primarily to the elimination of intergranular porosity by granules fracturing and deforming. At relatively high pressure regions (stage III), intragranule porosity elimination due to particle sliding and rearrangement is responsible for densification. The pressure level between stages II and III is defined as joining pressure[7] at which the coarse intergranular pores were eliminated. In general, the ease of deformation of granules as well as particle sliding/rearrangement are the principal factors in facilitating the compaction processing.

This study deals with the compaction behaviour of spray-dried silicon carbide powders. The powders were fabricated in this laboratory by a spray-drying technique.[8] Effects of relative humidity, organic

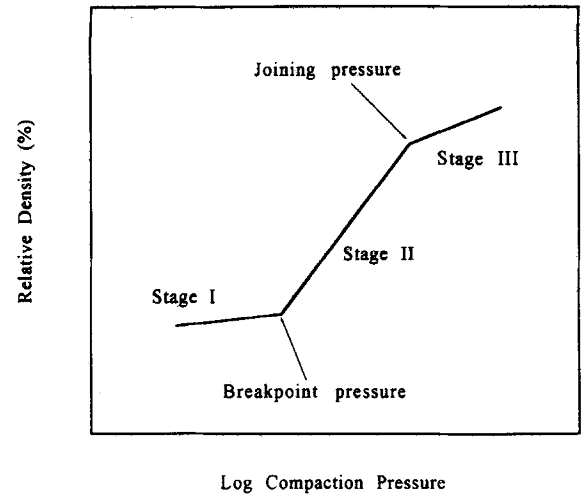

Fig. 1. Schematic density–compaction pressure curve.

phase content, temperature, and powder size and its distribution on the compaction behaviour of the SiC powders were investigated.

MATERIALS AND PROCEDURES

The spray-dried SiC powders were prepared from a slip containing 30 vol% SiC raw powders (Showa Denko, Japan), 0·5–3·0% organic phases, i.e. binder (acrylate-based polymeric material) and plasticizer (polyethylene glycol). The final organic phase content in the as-spray-dried granules was determined by TGA (Thermal Gravimetric Analysis, Netzsh). The powders that were used in the study are illustrated in Fig. 2, with particles nearly spherical in shape and with a geometric mean size of approximately 40 μm.

The effect of humidity on the compaction behaviour of the powders was examined by placing the powders under vacuum conditions for 48 h which was then designated as 'dry' powder (relative humidity < 5%), or storing in water-containing sealed vessel for 48 h as 'wet' powder (r.h. = 95%). Powders at ambient condition were designated as 'medium' powder (r.h. = 47%).

The compaction experiment was conducted by using a hardened steel die with a 23·21 mm × 23·21 mm cavity and punch set, lubricated with stearic acid to provide die wall lubrication. Seven ± 0·01 grams of the powder were poured into the die cavity, vibrated, and compacted at a pre-set pressure. After compaction, the specimen was removed and weighed (to 0·01 g and corrected for the organic phase content). The green densities of the specimens were measured using mercury porosimetry. Three to five specimens were used to determine the final density (within an error of ± 0·72%) of the compacts at a given pressure. The density–compaction behaviour can then be established at pressures ranging from 6·8 MPa to 154 MPa. To detect the elastic compression of the powder, the lower punch was fixed in the die and the movement of the upper punch was measured by an indicator to an accuracy of 0·01 mm. The height of the powder compact under a given pressure can then be determined in place following by calibrating with the elastic distortion of the die empty under the same pressure.

RESULTS AND DISCUSSION

Effect of relative humidity

Figure 3 shows the density–compaction pressure responses of the spray-dried silicon carbide powders for various relative humidities. The compact density increases with increasing relative humidity for a given pressure and this is similar to observations by Youshaw and Halloran[1] and Frey and Halloran[2] on the compaction of ferrite and alumina powders,

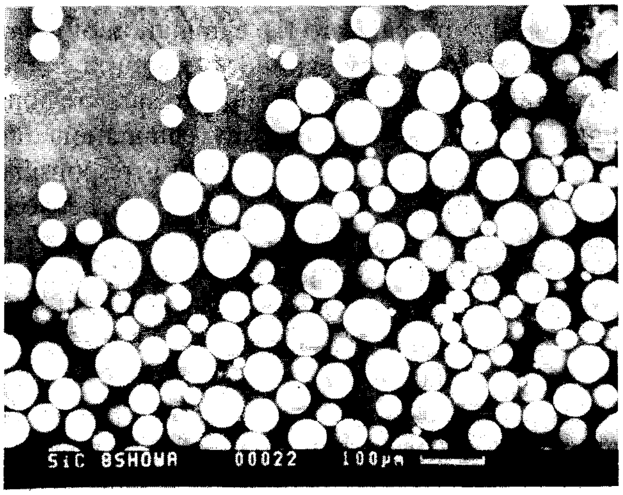

Fig 2. Morphology of the as-spray-dried silicon carbide powders (later designated as powder A).

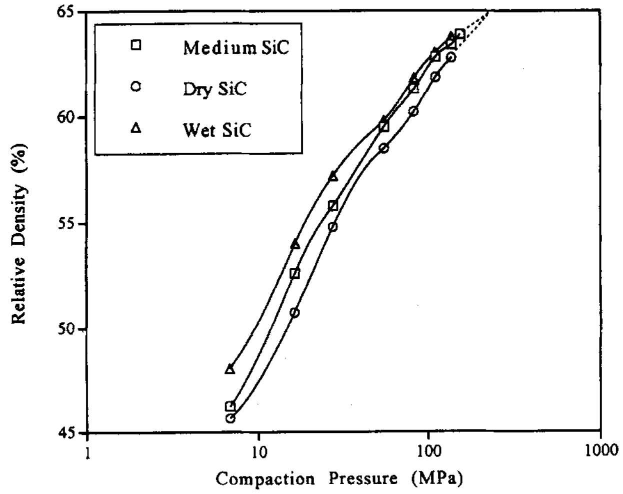

Fig. 3. The density–compaction pressure curves of the SiC powders under different humidities.

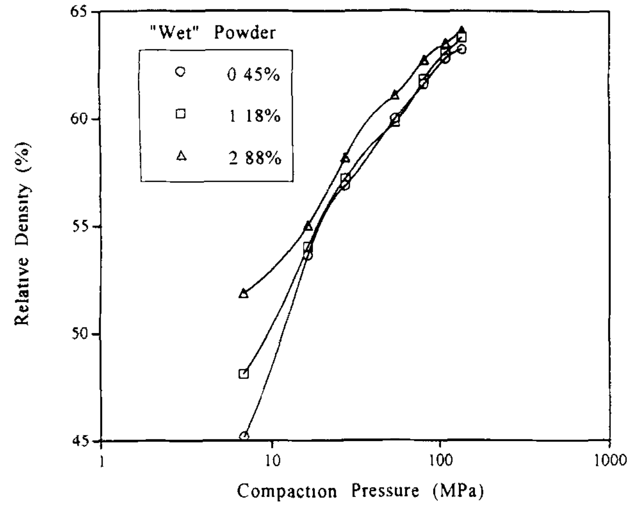

Fig. 4. Effect of organic phase content on the compaction behaviour of the SiC powders.

respectively. Figure 3 revealed that the difference in density becomes smaller at higher pressures, particularly for the 'medium' and 'wet' powders where approximately the same density is achieved at pressure > ~55 MPa (which may be regarded as the joining pressure), than at lower pressures. This suggests that the compaction mechanism for both the 'medium' and 'wet' powders is analogous when the pressure employed is greater than the joining pressure. On extrapolation of the density–compaction curves, an intercept pressure at ~200 MPa among these curves is attained. The joining pressure for both the 'medium' and 'wet' powders is obviously lower than that with the 'dry' powder indicating the ease of deformation of the granules for the former two powders. The 'dry' powders (having the lowest humidity) containing the organic phases may deform and/or fracture in a more brittle manner than the 'medium' and/or 'wet' powders (having higher humidity). The organic phases should become softer at higher relative humidity, causing the raw SiC particles to slide more easily than that in lower humidity condition, resulting in higher packing density.

Effect of organic phase content

The influence of organic phase content on density–compaction pressure behaviour is shown in Fig. 4 where the 'wet' powder is employed as a representative material. At lower pressure regions, the compact density increases with increasing organic phase content. The powders containing 0·45 and 1·18% organic phase show the same density at pressures greater than ~16·5 MPa. However, for 2·88% organic phase, a slightly higher value in density is maintained until at pressure of ~110 MPa, above which the density of the compacts appears to be independent of the organic phase content. As shown in Fig. 4, the compaction behaviour in terms of varying contents of organic phase suggests that at relatively low pressures, higher organic phase content promotes powder rearrangement, and hence removal of larger amounts of intergranular voids leads to a higher compact density. The organic phase, under moist conditions, acts more like a lubricant rather than a brittle solid and this phenomenon is expected to be more pronounced at the highest organic content. When further increasing the pressure, the difference in density among these powder compacts becomes small. This suggests that either the powder rearrangement mechanism is accelerated by increasing the pressure for the powders containing 0·45 and 1·18% organic phases or associated with the commencement of slight powder deformation and/or fracture, causing a rapid removal of intergranular porosity.

Effect of temperature

The influence of temperature on the compaction behaviour was investigated by using the powders containing 2·88% organic phase. The die and powders were equilibrated at 23, 50 and 70°C, prior to conducting the compaction test. The density–compaction pressure behaviours for various equilibrium temperatures are shown in Fig. 5. Increase of temperature decreases the compact density, the decrease being more pronounced at 70°C at lower pressure regions. In fact, the relative humidity at 70°C is relatively low and according to Fig. 3, a poor compaction is expected. However, at compaction > ~27 MPa, powder compacts at 70°C exhibit density slightly higher than those at other temperatures. Therefore, it is not possible to provide

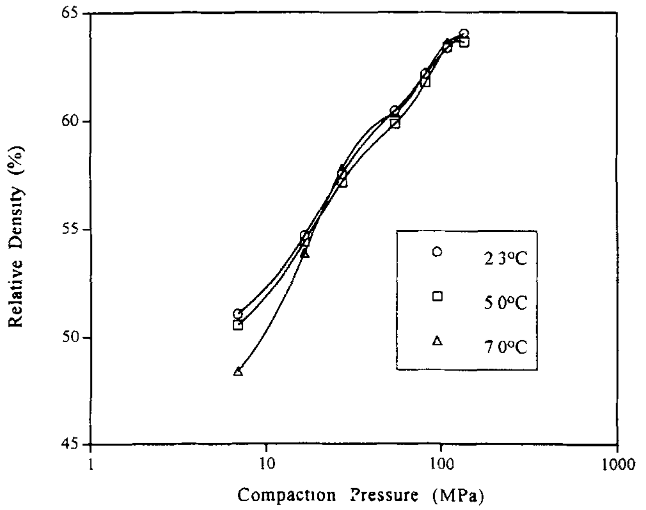

Fig. 5. Influence of powder temperature at 23°C, 50°C and 70°C on the density–compaction pressure relations.

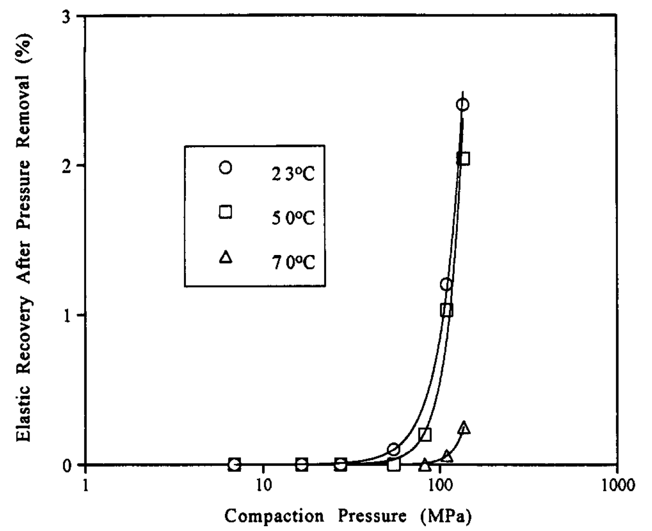

Fig. 6. Elastic recovery of the compact SiC powder after removal of the compression force.

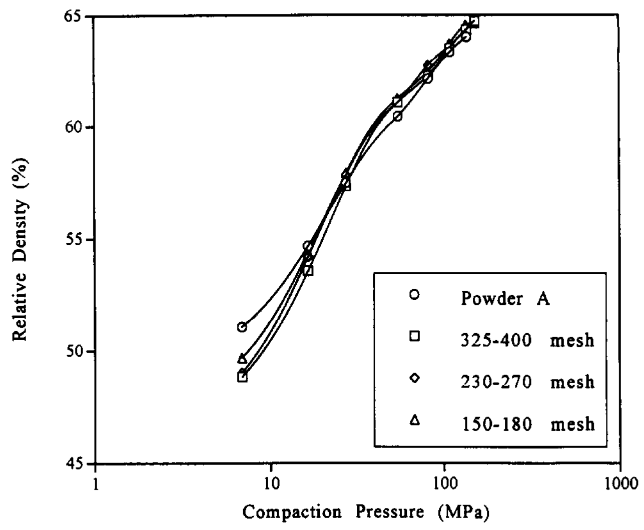

Fig. 7. Effect of powder size on the compaction behaviour of the SiC granules.

a satisfactory explanation by considering only the humidity effect. Differential thermal analysis (Netzsh) revealed that the glass transition temperature (T_g) of the organic phase is approximately 60°C. This finding may offer a reasonable explanation that the powder rearrangement is retarded by poorer powder flow during low-pressure compaction (stage I). At higher pressures, the powders were fractured and deformed plastically and the viscous effect of the organic phase may facilitate one particle sliding over the other. This promotes the removal of intragranular porosity and provides a better packing configuration, resulting in better compaction density. A similar trend was reported on the influence of T_g on the compaction of Al_2O_3 granules with polyvinyl alcohol as organic phase.[9]

Figure 6 shows that elastic recovery (~2%) of the compressed SiC powders was observed after removal of compaction pressure at ~100 MPa at 23 and 50°C. However, such elastic recovery is minimized below ~0.3% above the glass transition temperature. Such a small amount of recovery at 70°C is believed to be due solely to the elastic springback of the compressed SiC particles. Figure 6 reveals the importance of the elastic nature of the organic phase for quality control in industrial operations because a sufficient level of elastic recovery from either organic phase, or SiC, or both may usually cause serious cracking and delamination within the compressed bodies, and in consequence causes damage in final sintered products.

Effect of powder size

The spray-dried powders were sieved into three different size fractions, namely, 325–400 mesh, 230–270 mesh, and 150–180 mesh, corresponding to mean powder sizes of approximately 40 μm, 60 μm, and 100 μm, respectively. The influence of powder size on compaction behaviour is shown in Fig. 7. The green compacts usually exhibited a lower value of density for the coarser powders than for the finer ones at compaction pressures below approximately 28 MPa. Since at lower pressure regions, the compaction behaviour is limited to powder rearrangement and/or partial elimination of intergranular porosity, the coarser powders will usually provide a lower fill density corresponding to a higher intergranular porosity compared to that of the finer powders, resulting in lower compact density. At pressures greater than ~28 MPa, which may correspond to a stage II compaction (Fig. 1), the powder size appeared to have little or no influence on the resulting compact density, which suggests that the post-yield compaction is hardly affected by powder size. These phenomena were also observed in the compaction of alumina granules by Frey and Halloran.[2]

Effect of powder size distribution

Powders of uniform size are seldom used in commercial pressing operations; on the contrary, powders

Table 1. Powder size distribution for powders A and B

Powder size (mesh)	Powder A (wt%)	Powder B (wt%)
100-120	0·87	0·9
120-150	0·96	0·44
150-180	2·99	22·8
180-200	0·23	5·53
200-230	20·4	19·7
230-270	35·52	17·6
270-325	14·44	7·4
325-400	0·086	0·18
+400	24·17	25·4

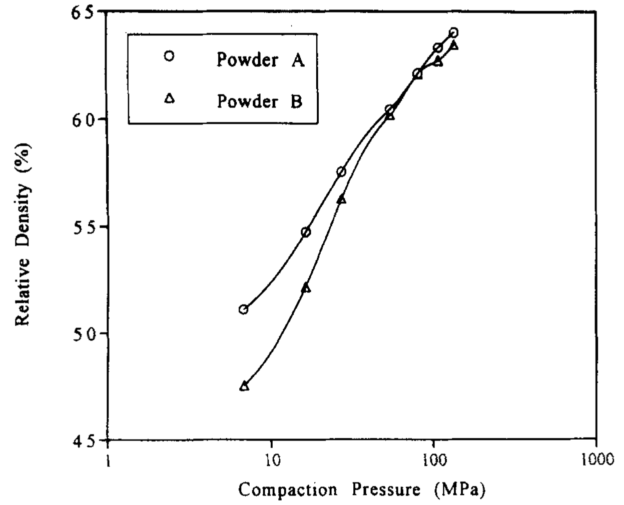

Fig. 8. Comparison of the density–pressure diagram for the two SiC powders with different powder size distribution.

Fig. 10. SEM photo shows some of undeforming and unfracturing smaller powders at 16·5 MPa.

of broader size distribution are frequently encountered in industry production. Two kinds of spray-dried silicon carbide powders, namely powder A and powder B, were designed to elucidate the influence of powder size distribution on the compaction behaviour. Both powders contain the same quantity of organic phase, i.e. 2·88 wt%. The powder size distributions of both the powders are given in Table 1. Powder A has a geometric mean size of $\sim$40 μm and powder B $\sim$65μm.

The density–compaction pressure behaviour of the powders is shown in Fig. 8. The green compacts derived from powder A usually exhibited a higher green density than those from powder B at pressures below approximately 55 MPa, and both compacts showed roughly similar densities at pressures of 55–83 MPa. Further increase of the compaction pressure above $\sim$83 MPa slightly reduced the green density of the compacts derived from powder B. Because of limitations of the sieving apparatus, the powders below $\sim$37 μm (i.e. + 400 mesh) cannot be further classified into different size fractions. However, from Table 1, powder A has a smaller quantity, i.e. $\sim$24·1%, below 37 μm than powder B, i.e. $\sim$25·4%. An observation of powder B (Fig. 9) shows that it contains a large quantity of small granules (some of them are only $\sim$10 μm in diameter) and a few of the granules contain large craters. These powder characteristics may partially explain the compaction behaviour illustrated in Fig. 8. The lower compaction density for powder B at low pressure regions may be caused by un-deforming and/or slightly deforming of the small granules which fit loosely in the interstices among larger granules, as shown representatively in Figs 10 and 11. This implies that a combination of two or more compaction mechanisms may occur simultaneously but to different extents. At sufficiently high compressive pressures, e.g. $\geq$ 83 MPa, the lower green density of the powder-B-derived compacts may be caused by the presence of powder imperfections such as craters (denoted with arrows in Fig. 9) which may restrict the compaction behaviour as observed by Lukasiewicz and Reed[7] who found that some craters may exist even under compaction pressures greater than 100 MPa.

Fig. 9. Scanning electron micrograph of powder B, showing the presence of craters.

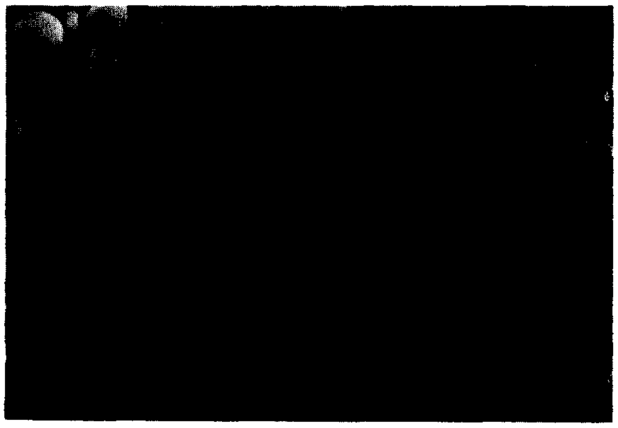

Fig. 11. Some granules may keep only slightly deforming under compaction pressure as high as 55 MPa (8,000 psi).

CONCLUDING REMARKS

Compaction behaviours of spray-dried silicon carbide powders were investigated using relative humidity, powder temperature, organic phase content, powder size, and powder size distribution as variables. At higher compaction pressures, e.g. ≥ 55 MPa, the influence of humidity, organic phase content, temperature, and powder size is considerably reduced. However, they are significant at lower pressure regions. The compaction behaviour of the SiC powders with wide powder size distribution is believed to be made up of a combination of two or more mechanisms, particularly for powders containing sufficient quantities of smaller granules, e.g. ≤ ~10 μm in diameter. To avoid damage of the compacts due to compressed elastic recovery, a compaction temperature greater than 60°C is necessary for the organic phase used in this investigation.

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REFERENCES

1. YOUSHAW, R. A. & HALLORAN, J. W., Compaction of spray-dried powders. *Am. Ceram. Soc. Bull.*, **61**(2) (1982) 227–30.
2. FREY, R. G. & HALLORAN, J. W., Compaction behaviour of spray-dried alumina. *J. Am. Ceram. Soc.*, **67**(3) (1984) 199–201.
3. MESSING, G. L., MARKHOFF, C. J. & McCOY, L. G., Characterization of ceramic powder compaction. *Am. Ceram. Soc. Bull.*, **61**(8) (1982) 857–60.
4. LEISER, D. B. & WHITTMORE, O. J., Jr., Compaction behavior of ceramic particles. *Am. Ceram. Soc. Bull.*, **49**(8) (1970) 714–17.
5. HUFFINE, C. L. & BONILLA, C. F., Particle-size effects in the compression of powders. *AIChE J.*, **8**(4) (1960) 490–3.
6. STEPHEN, R. G. & RILEY, F. L., The compaction behavior of oxidised powder. *J. Europ. Ceram. Soc.*, **9** (1992) 301–7.
7. LUKASIEWICZ, S. L. & REED, J. S., Character and compaction response of spray-dried agglomerates. *Am. Ceram. Soc. Bull.*, **57**(9) (1978) 798–805.
8. Internal Report, Material Research Laboratories, ITRI.
9. DiMILIA, R. A. & REED, J. S., Dependence of compaction on the glass transition temperature of the binder phase. *Am. Ceram. Soc. Bull.*, **62**(4) (1983) 484–8.