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Research Article

Experimental Investigation on Flowability and Compaction Behavior of Spray Granulated Submicron Alumina Granules

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Aqueous slurry with various solid loadings (up to 40 wt%) of alumina powder (D_{50} = 300 nm) with suitable rheological properties were spray dried into granules. Solid loading and feed rate of the slurry are found to have a prominent effect on the shape and size distribution of granules. Powder flow measurements exhibited a cohesive index of 28.45 signifying an extremely cohesive flow due to high surface area and irregular morphology. Finer sizes though it offers high geometrical surface area it leads to more surface contacts and hence, high interparticle friction. Spherical morphology achieved through optimum spray drying parameters significantly reduced the cohesive index to 6.45 indicating free flow behavior. Compaction studies of the spray-dried granules and corresponding plot of relative density versus compaction pressure revealed an agglomerate strength of 500 MPa followed by a plateau-like behavior reaching a maximum in the relative density of 59%-60% of the theoretical values.

1. Introduction

Flow properties of the powders are dictated by the collective forces acting on individual particle such as vander waals and electrostatic forces, surface tension, interlocking as well as friction. Compaction process of ceramic powders basically involves die filing and particle rearrangement under applied stress followed by brittle fracture and bonding between the particles [1–6]. Further, the removal of the applied pressure and ejection of the compact completes the compaction process. Flow variables such as energy required to ensure an acceptable flow, sensitivity to flow rate as consequence of the previous collective forces, and distribution of the particles under applied stress play a major role in determining the quality of the formed compact. Flow properties of the fluids and pastes are characterized in terms of their rheological properties but similar treatment cannot be applied to the powders. Several experimental techniques have been developed to determine the flow behaviors of the powders. The flow properties are generally characterized by physical measures such as angle of repose, flow and shear based methods and even correlated with tap density and green strength of the compact [7–9]. Flowability of powder is a complex parameter that cannot be described with a single number and there is no universal model in existence to predict powder flow behaviour in every situation. Particulate flow characteristics are complex and flow properties are a combined effect of physical and environmental variables. Hence, flowability is a combination of physical property of the powders and generally correlated with the fundamental measurable characteristics [10–12].

Particle size and distribution, shape, surface area, porosity, particle density, and surface texture are few of fundamental measurable properties of the powder and relate to the flow behavior. For example, if the particle size is small and with high surface to volume ratio, the flow is restricted due to the cohesive force between the particles and flow properties can be improved by engineering the optimum size of granules. In the present study, alumina spray granulated to achieve a variety of granule morphologies. Flow measurements were carried out with selected granules for as-received and spray granulated powder that exhibited the best spherical granules (granules obtained with solid loading 40 wt%). Parameters such as cohesion index, compaction coefficient, and powder flow speed dependency were carried out using a powder flow analyzer attached to a Texture Analyzer [13]. Flow properties

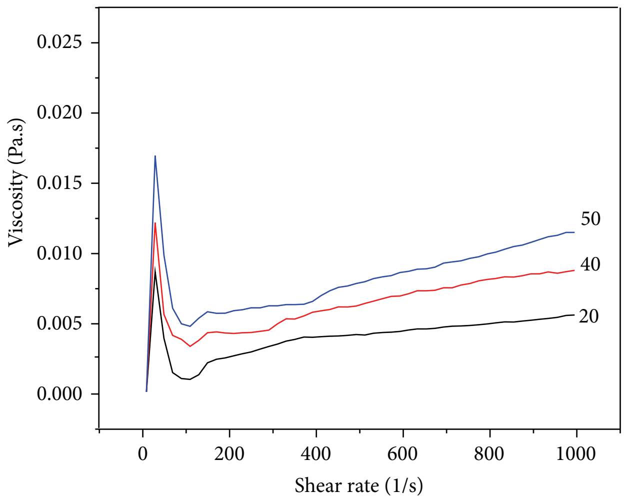

FIGURE 1: Plots of viscosity with shear rates.

were correlated with the basic properties of the granules and compaction behavior of the granules was also investigated as reported in this study. Compaction studies were also carried out for spray-dried granule in order to establish a quantitative relationship between the flow behavior and the resultant compact density.

2. Materials and Methods

Commercially available alumina powder (AHPA, Ceralox, USA) with the specifications (provided by the supplier) 99.96% purity, surface area 8 m^2/gm, and average particle size D_{50} = 300 nm was used in this study. The powder was made into an aqueous slurry in distilled water using 1 wt% Darvan 821A as a dispersant and 2 wt% PVA as the binder. The suspension was then milled for 2 hrs in polypropylene bottles in a pot jar mill using alumina balls of 2 mm diameter and 1 : 1 charge to balls ratio. A solid loading of alumina in the range of 20, 40, and 50 wt% was achieved through the previous milling parameters. Rheological behavior of the slurries was measured at varying shear rates (MCR 51, Anton Paar, Austria) to determine the flow properties. A laboratory spray drier (BUCHI B 290/295, Switzerland) was employed in this study which consists of a peristaltic pump that feeds the slurry into the nozzle which is located at the top of the drying chamber and produces a fine spray. The hot air entry was made in counter current with the slurry feed. Granules are collected at the bottom of the chamber and further separated by the cyclone separators. Operating parameters such as entry air temperatures were kept at maximum of 210°C and accordingly the exit temperatures at equilibrium were found to be 145 ± 2°C and 148 ± 2°C for 20% and 40% solid loading, respectively. Two fluid atomization nozzle designs with a nozzle diameter of 1.4 mm and slurry feed rate of 3.75 to 7.5 mL/minute are employed to produce various spray granulated (SG) samples.

Cohesion index measurements were carried out by the powder flow analyzer attached to a Texture Analyzer (Stable Micro System, UK). As-received powder and the best spherical granules were evaluated using the previous equipment. As per the standard procedure, 50 gm of the powder was used in each case of measurements. A programmed motion of the blade in the cylinder confining the granules ensures flow modes of compression while moving downwards in clockwise direction and slicing while moving anticlockwise. As the blade moves up clockwise the lifting of the material occurs. The detailed procedure of the measurement is described elsewhere [14].

Uniaxial dry-pressing is carried out with as-received powder and spray-dried granule in order to establish a quantitative relationship between the flow behavior, compaction pressure, and the resultant compact density. Also, 5 gm of the powder or granule was placed in the die in each case and displacement of the upper punch was used to calculate the change in volume of the powder compact as a function of applied pressure. Pressure-density curve was generated based on the load versus displacement curve recorded during compaction of the granules.

3. Results and Discussion

3.1. Rheological Characterization. Figure 1 presents the viscosity measurements with varying shear rates of alumina slurries with 20, 40, and 50 wt% of solid loading.

It is evident that there is an overshoot in the viscosity curve which is increasing during the initial shear which can be attributed to the initial yield stress. The overshoot is evident in 20 to 40 wt% but becomes more prominent in the case of 50 wt% indicating the requirement of higher extent external force to initiate the flow. When the solid loading is increased with alumina particles of average particle size (D_{50}) of 300 nm interparticle forces become important in controlling the rheological responses as is evident from the high initial yield stress values. However, for optimal spray drying, a high solid loading with low viscosity is preferable. It is evident that in the present study, the rheological behavior is not affected very much even with 50% of solid loading except the initial overshoot. This negligible increase in viscosity values with solid loading can be attributed to the dispersant-binder compatibility (Darvan 821A and PVA) which necessarily maintains optimal slurry rheology while spray granulation.

3.2. Spray Granulation. Spray drying conditions and parameters are shown in Table 1. Scanning electron micrographs of as-received (Al-a) powder and spray-dried granules with 20 wt% solid loading (SG-20) under various feed rates of 3.75, 5, 6.25, and 7.5 mL/min are shown in Figures 2(b)–2(h). For the sake of comparison, spray-dried granules with 40 wt% solid loading (SG-40) under various feed rates of 3.75 and 7.5 mL/min are shown in Figures 2(i) and 2(k). It is evident that as-received powder has shown an irregular morphology.

Spray drying has resulted in agglomeration of particle irrespective of the feed rate and solid loading; however, the shape of the granules and surface texture is found to be a

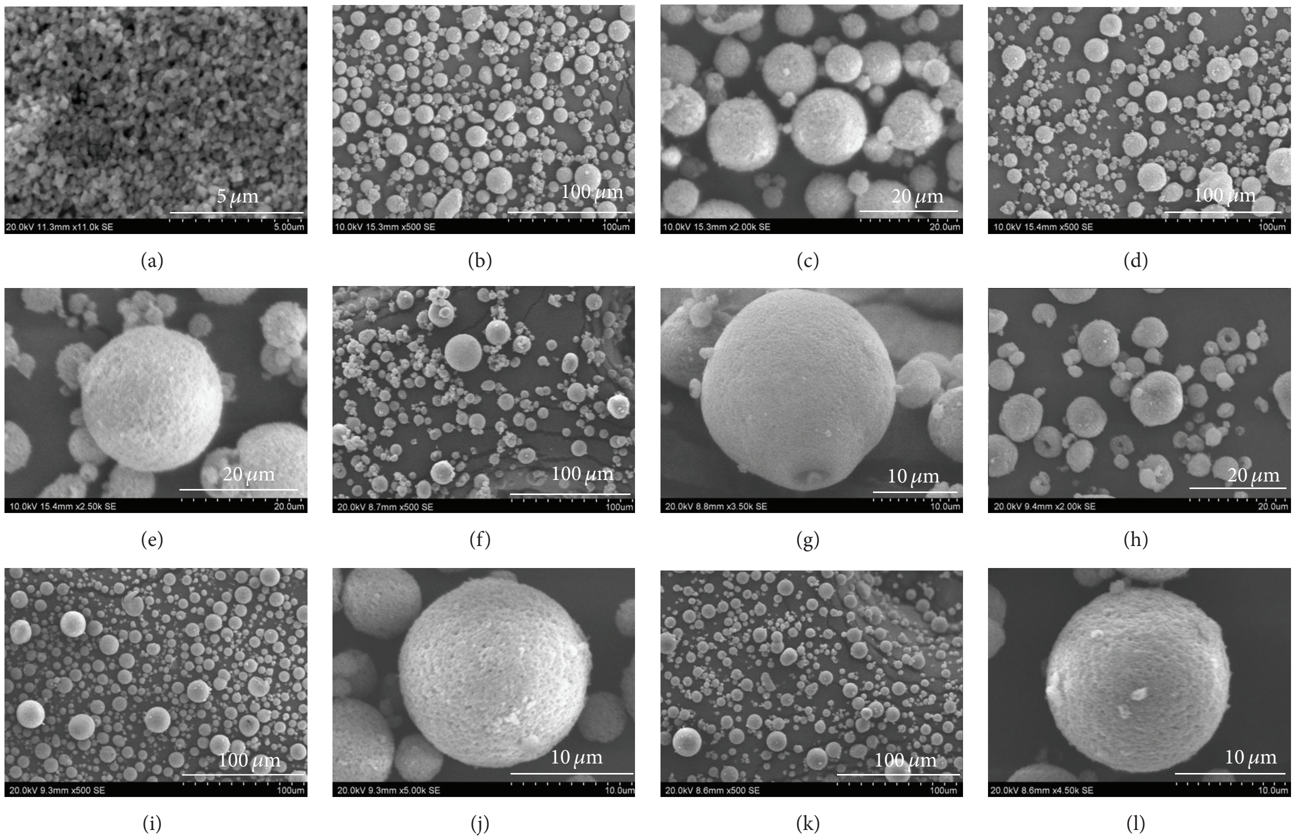

Figure 2: Micrographs of Al (a) and SG-20 ((b)–(h)) along with magnified images. Micrographs of SG-40 ((i)–(l)) along with magnified images.

Table 1: Slurry and spray drying variables.

Sample ID	Inlet air temperature (°C)	Outlet air temperature (°C)	Solid loading (wt%)	Atomization nozzle dia (mm)	Atomization air pressure (bar)	Feed rate (mL/min)
SG-20						
b	210	145	20	1.4	6	3.75
d	210	145	20	1.4	6	5.0
f	210	145	20	1.4	6	6.25
h	210	145	20	1.4	6	7.5
SG-40						
i	210	148	40	1.4	6	3.75
k	210	148	40	1.4	6	7.5

Slurries with solid loading of 50 wt% could not be sprayed due to pump limitation.

strong function of solid loading and feed rate. For SG-20, a clear effect on shape of the granules is evident from the increase in the feed rate of the slurry. Granules are perfectly spherical up to the feed rate of 5 mL/min. However, the granules lost their sphericity with increase in feed rate beyond 5 mL/min. The granules resulted in dumb bell shape even with the formation of cater in the middle of granule in Figure 2 SG-20 (g) and (h). On the contrary, with the SG-40 slurry samples the granules were spherical irrespective of the feed rate. Moreover, spray-dried granules are porous composites consisting of original submicron sized particle contained in the binder matrix and surface tension is one of the key properties of slurry that determines the shape of the granule. During spray drying the well-dispersed particles with low viscosity are in fact mobile in the dispersing medium which migrate fast to the surface and result in the formation of hollow granules with the shell. On the other hand, in the case of slurry with high viscosity, the particle has low mobility in the medium and remains as solid granules. The sphericity of the granules originated from slurry with solid loading of 40% irrespective of the feed rate can be attributed to the relatively high viscosity and the resulting surface tension

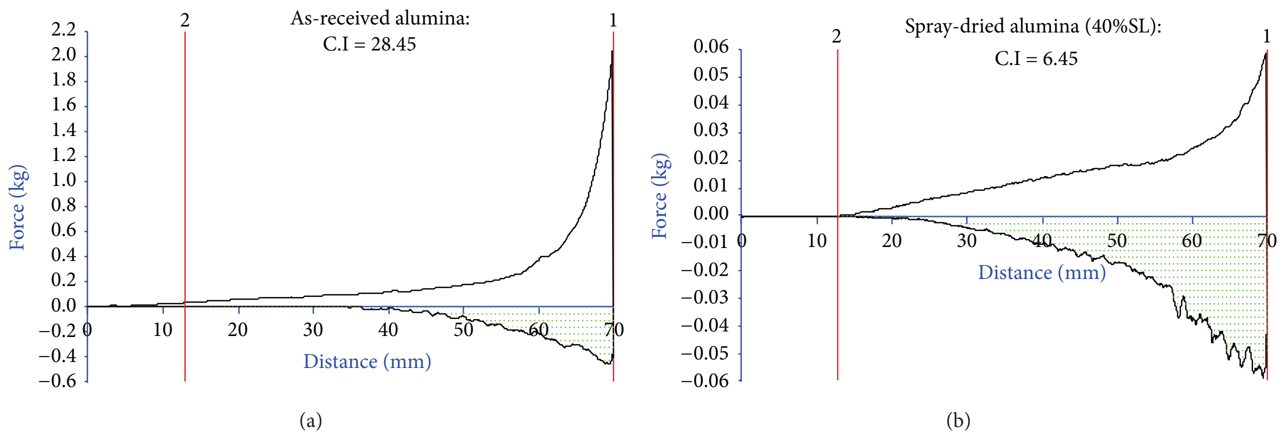

FIGURE 3: Force versus distance curves for alumina and SG-40 spray-dried granules along with cohesion index.

TABLE 2: Effect of feed rate on granule size and distribution.

Sample ID	Feed rate (mL/min)	Average size of the granule (μm)	Size distribution (μm)
SG-20			
b	3.75	20	10–30
d	5	18	15–30
f	6.25	16	10–20
h	7.5	15	10–20
SG-40			
i	3.75	12	10–20
k	7.5	15	10–20

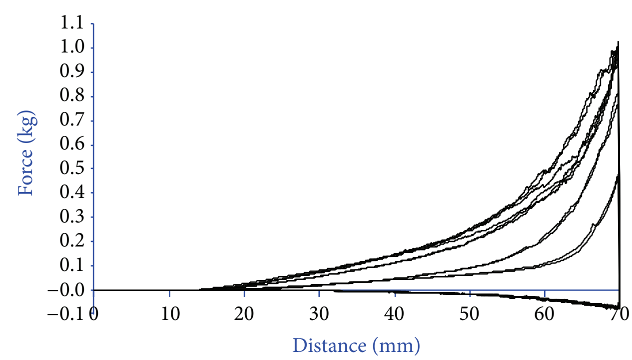

FIGURE 4: Plot of force versus displacement curves for spray-dried alumina (SG40).

that retain the matrix in spherical form while drying in the chamber.

Further, the granule size and distribution are shown in Figure 2. It is also evident from Table 2 that size and distribution of the granules mainly depend on the solid loading and feed rate of the slurry. Further, the magnified images of surface of the spray-dried granules in all cases exhibited high porosity signifying its soft nature. Soft nature of the granules is very important for compaction processing as these need to be completely broken down while pressing. However, soft granules may undergo fracture or deformation causing negative effects on flowability leading to problems with handling and mold filling.

3.3. Cohesion Index (C.I) Measurement. Cohesion index measurements were carried out for as-received alumina (Figure 2: Al (a)) and the best spherical granules (Figure 2: SG-40 (i)). Plots of force versus distance for both samples are shown in Figures 3(a) and 3(b). Cohesion index is regarded as the ratio of cohesion coefficient and the weight of the sample evaluated and is calculated by integrating the negative areas under the curve using standard software provided with the equipment. Though the curves are recorded for several cycles for the sake of simplicity a typical force versus distance curve of as-received alumina and SG-40 (i) samples is only shown in Figure 3 along with the cohesion index.

Table 3 shows the reference flowability categorization based on cohesion index values. It is very interesting to note that a cohesion index value of 28.45 indicating an extremely cohesive flow behavior for as-received alumina could be modified significantly by spray drying. A cohesive index of 6.45 for SG-40 (i) granules indicates a free flowing behavior. Transformation of very cohesive flow behavior to free flowing can be attributed to the narrow distribution of granules in combination with the spherical morphology as is evident from the micrographs depicted in Figure 2 SG-40 (i), respectively.

3.4. Powder Flow Analysis of Granules. A typical plot of force versus displacement curves is recorded for SG-40 during Powder Flow Speed Dependency (PFSD) test as shown in Figure 4.

Alumina powder without spray granulation could not be evaluated because of the overloading of the system indicating high resistance to the flow with increasing shear rate. However, the spray-dried powder behaved totally different as there is a decrease in the compaction coefficient with the increase in test speed. This indicates a decreasing resistance to the flow

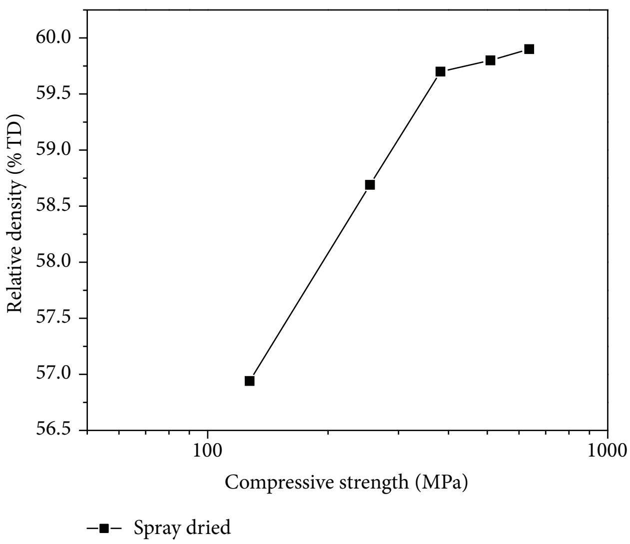

Figure 5: Plot of relative density versus compaction pressure.

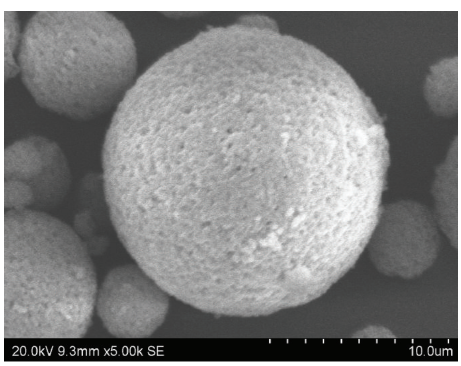

Figure 6: Micrographs of SG-40 granules magnified showing the inter-particle porosity.

with increase in solid loading in case of spray-dried powders and therefore exhibits flow speed dependence.

3.5. Evaluation of Compressibility Curve. In order to have a better understanding on behavior of spray-dried granules under uniaxial compaction compressibility curves were recorded for the granules (SG-40 (i)).

Compressibility curves of the spray-dried granules are shown in Figure 5. It is evident that there is a distinct slope change in the plot of relative density versus compressive strength. Due to the free flowing nature of the granules as shown by the powder flow analysis, rearrangements to fill the intergranular voids are expected to take place at very low pressures. A steep increase in densification rate marks the reach of agglomerate strength (500 MPa) and breakage of individual highly porous (Figure 6) soft granules with high porosity. As the compaction pressure increases, porosity within the granules is continuously eliminated and exceeds the yield strength of all the granules. It is evident that the compressibility curve almost became a plateau and the relative density of compact would be equal to the density of a bulk with further increase of compaction pressures reaching the maximum relative density without lamination defects.

Table 3: Reference flow ability categorization based on cohesion index.

Sr. no.	Cohesion index	Flow behavior
1	19+	Extremely cohesive
2	16–19	Very cohesive
3	14–16	Cohesive
4	<14	Free flowing

4. Conclusion

High purity alumina powders with D_{50} = 300 nm were made into aqueous slurries with solid loadings of 20, 40, and 50 wt% with optimum dispersant-binder concentrations (Darvan 821A and PVA) which exhibited suitable rheological properties during spray granulation.

Solid loading and feed rate of the slurry are found to have a prominent effect on the shape and size distribution of granules. For SG-20 granules are perfectly spherical up to the feed rate of 5 mL/min and the granules lost their sphericity with increase in feed rate beyond 5 mL/min. However, for SG-40 slurry samples the granules were spherical irrespective of the feed rate which could be attributed to the relatively high viscosity and the resulting surface tension that retain the matrix in spherical form while drying in the chamber.

Average particle size of D_{50} = 300 nm with high surface area and irregular morphology results in more surface contacts resulting in frictional forces which resist the flow exhibiting a high cohesive index of 28.45. Cohesive index of 6.45 of SG-40 (i) granules is due to spherical morphology achieved through optimum spray drying parameters.

Compressibility curve of the SG-40 (i) granules exhibited a steep increase in densification rate after crossing the rearrangement stage reaching 500 MPa agglomerate strength and a plateau where the relative density of compact would be equal to the density of the bulk limiting the relative density values.

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