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Rapid Communication

Use of First Derivative of Displacement *vs.* Force Profiles to Determine Deformation Behavior of Compressed Powders

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Abstract. Displacement (D) *vs.* force (F) profiles obtained during compaction of powders have been reported by several researchers. These profiles are usually used to obtain mechanical energies associated with the compaction of powders. In this work, we obtained displacement–force data associated with the compression of six powders; Avicel PH101, Avicel PH301, pregelatinized corn starch, anhydrous lactose, dicalcium phosphate, and mannitol. The first three powders are known to deform predominantly by plastic behavior while the later ones are known to deform predominantly by brittle fracture. Displacement–force data was utilized to perform in-die Heckel analysis and to calculate the first derivative (dD/dF) of displacement–force plots. First derivative results were then plotted against mean force (F′) at each point and against 1/F′ at compression forces between 1 and 20 kN. Results of the in-die Heckle analysis are in very good agreement with the known deformation behavior of the compressed materials. First derivative plots show that materials that deform predominantly by plastic behavior have first derivative values (0.0006–0.0016 mm/ N) larger than those of brittle materials (0.0004 mm/N). Moreover, when dD/dF is plotted against 1/F′ for each powder, a linear correlation can be obtained (R^2=>0.98). The slopes of the dD/dF *vs.* 1/F′ plots for plastically deforming materials are relatively larger than those for materials that deform by brittle behavior. It is concluded that first derivative plots of displacement–force profiles can be used to determine deformation behavior of powders.

KEY WORDS: compression; deformation; first derivative.

INTRODUCTION

Displacement *vs.* force profiles of powder compression experiments have been firstly reported by several researchers (1–4). These profiles have since been used to obtain certain information regarding the compaction behavior of powders. These researchers were among the first who utilized force–displacement profiles to obtain mechanical energy estimations. De Blaey *et al.* (1) used force–displacement profiles to obtain coefficients of lubrication and used these values to compare die wall lubrication and lubrication through incorporation of lubricant in the granulation. Rasenack *et al.* (5) utilized force–displacement profiles along with resulting compact properties to calculate a comparative factor (T factor) which was used to compare tableting behavior of different substances. Accordingly, substances with relatively high calculated T factor are suggested to have good tableting behavior. Heckel (6) suggested an equation that describes the relation between compact porosity and applied pressure which can be used for prediction of deformation mechanisms of powders. Moreover, displacement–force data can be used to construct the aforementioned relation and obtain what is known as the "in-die" Heckel analysis. Further attempts to use displacement–force profiles to predict the deformation mechanism of powders have been reported by Antikainen *et al.* (7). Accordingly, displacement–force profiles were utilized to predict the extent of plastic flow, fragmentation, and elastic recovery of powders. Their method principally relied on the fact that near maximum displacement the upper punch moves very slowly and plastic materials require time to deform. However, for materials that deform predominantly by fragmentation the displacement at maximum pressure depends only on pressure and does not change with time.

In this work, and for the first time, the first derivative of displacement *vs.* force plots was analyzed to obtain first derivative plots. These plots were used for differentiating plastic and brittle deformations of powders.

MATERIALS AND METHODS

All powders were used as received from suppliers. Compacts were obtained by using an Instron Universal Testing System with a 50-kN load cell (Model 5569, Instron Corp, Norwood, MA, USA). Powders of 300±2 mg were filled into a 10-mm die and were compressed and decompressed at 50 mm/min up to a specified compression load of 20 kN.

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Flat-faced punches were used. The punches and die were lubricated with a 5% (*w*/*v*) magnesium stearate suspension in methanol before each set of triplicates and allowed to dry. All powders and compacts were stored at ambient conditions. Temperature and RH% were recorded (typically 20–22°C, and 50–60% RH).

A total of six powders were chosen; Avicel PH 101 and Avicel PH 301 were obtained from FMC Corporation (Philadelphia, PA, USA), Pregelatinized corn starch from Colorcon (West Point, PA, USA), anhydrous lactose from Foremost Farms (Baraboo, WI, USA), mannitol from Roquette (Lestrem, France), and dicalcium phosphate (dibasic calcium phosphate dihydrate) from Rhodia Inc. (Cranbury, NJ, USA). The first three materials are known to deform predominantly by plastic behavior under pressure (8). However, the last three materials are known to deform predominantly by brittle fracture under pressure (8–12). In-die, Heckel analysis (2) was performed on all powders. In addition, the first derivative (dD/dF) at each point of the force–displacement profile was determined according to the following equation:

$$\frac{\mathrm{dD}}{\mathrm{dF}} \cong \frac{\Delta D}{\Delta F} = \frac{(D_2 - D_1)}{(F_2 - F_1)} \tag{1}$$

Where, D_2 is the registered displacement at force F_2. D_1 is the registered displacement at force F_1. The derivative values were determined and plotted against the average force (F′) at each point or plotted against 1/F′ at each point for analysis purposes.

RESULTS AND DISCUSSION

Figure 1 shows displacement *vs.* force profiles for the compression phases of the studied powders. It can be seen from the figure that the profiles for Avicel PH101, Avicel PH301, and pregelatized corn starch are shifted upwards compared to the rest of powders. Such shift is due to further displacement resulting in greater potential in the powder bed to yield to applied compression force. This is consistent with the fact that materials that deform predominantly by plastic flow yield more to compression force. Table I shows a summary of in-die Heckel analysis for the tested powders. Avicel PH101, Avicel PH301, and pregelatinized corn starch have smaller yield pressure values when compared to anhydrous lactose, dicalcium phosphate, and mannitol. This is consistent with the fact that Avicel PH101, Avicel PH301, and pregelatinized corn starch deform predominantly by plastic behavior under pressure. However, anhydrous lactose, dicalcium phosphate, and mannitol have larger yield pressure values, which indicates a predominantly brittle behavior under pressure. Note that the force–displacement profile for starch appears to be closer to that of anhydrous lactose, dicalcium phosphate, and mannitol (brittle materials) rather than plastic likely due to the viscoelastic nature of starch.

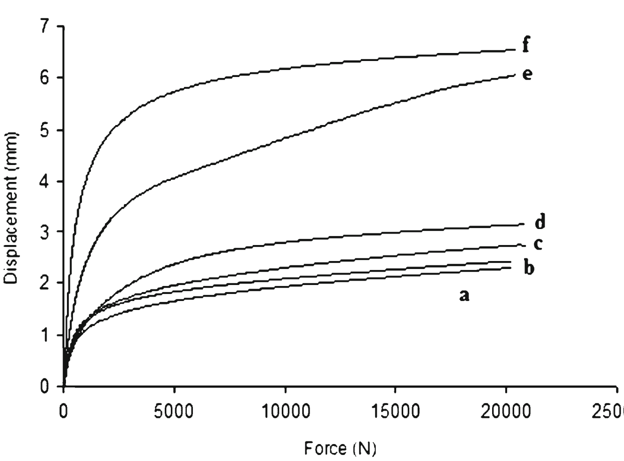

Fig. 1. Displacement *vs.* force profiles for *a* mannitol and *b* dicalcium phosphate, *c* anhydrous lactose, *d* pregelatinized corn starch, *e* Avicel PH301, *f* Avicel PH101

Table I. Summary of Heckel Plot Parameters for Tested Materials (n=3)

Material	Average yield pressure (MPa)	Standard deviation	Average intercept	Standard deviation	R^2
Avicel PH101	55.9	0.73	0.61	0.003	>0.998
Avicel PH301	54.9	1.05	0.56	<0.001	=0.995
Pregelatinized corn starch	48.7	0.60	0.72	0.003	>0.998
Anhydrous lactose	147.5	7.18	1.19	0.010	>0.999
Dicalcium phosphate	268.9	1.00	1.28	0.004	>0.999
Mannitol	92.7	4.00	1.41	0.011	>0.999

Figure 2 shows a plot of dD/dF plotted against F′ for Avicel PH101, Avicel PH301, and pregelatinized corn starch at compression forces between 1 and 20 kN. It can be seen that dD/dF has values between 0.0006 and 0.0016 mm/N at 1 kN for these powders. Moreover, Fig. 3 shows dD/dF plotted against F′ for anhydrous lactose, dicalcium phosphate, and mannitol at compression forces between 1 and 20 kN. It can be seen that dD/dF has values for these materials of about 0.0004 mm/N at 1 kN. Comparison of dD/dF values indicates that materials that deform predominantly by plastic behavior (Avicel PH101, Avicel PH301, and pregelatinized corn starch) have relatively larger dD/dF values at a compression force of 1 kN when compared to materials that deform predominantly by brittle fracture (anhydrous lactose, dicalcium phosphate, and mannitol). This is in agreement with the fact that plastic materials yield more to applied pressure. In addition, Figs. 2 and 3 show that dD/dF for each material declines sharply at small to moderate compression forces and plateaus (approaches zero) at high compression forces (>15 kN). This indicates negligible movement of the upper punch at high compression forces.

Figure 4 shows the relationship between dD/dF and 1/F′ for all of the tested materials. It can be seen that a linear relationship between dD/dF and 1/F′ can be established (R^2= >0.98) in all figures. Moreover, the values of the slopes of the fitted lines are different. Table II lists a summary of the slope, intercept, and R^2 values of the fitted lines. It can be clearly seen that materials, which deform predominantly plastically (Avicel PH101, Avicel PH301, and pregelatinized corn starch) have larger slope values (0.69–1.42 mm) when compared to materials that deform predominantly with brittle fracture (anhydrous lactose, dicalcium phosphate, and

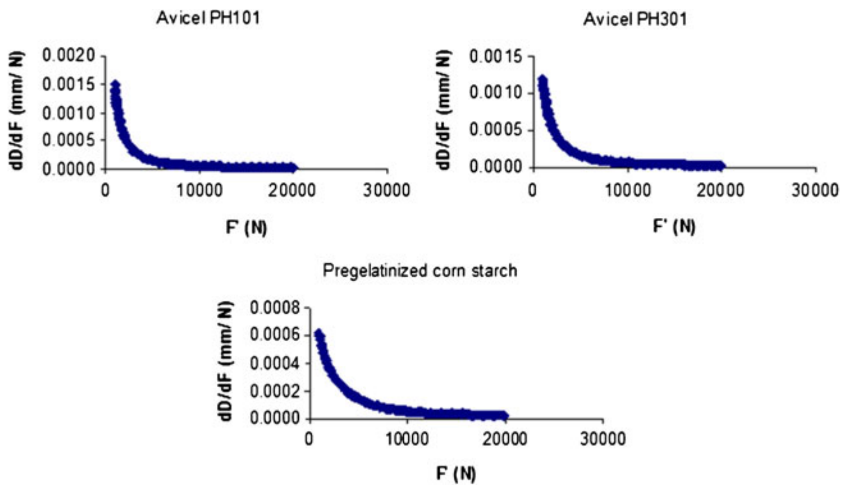

Fig. 2. Relation between first derivative and compression force for Avicel PH101, Avicel PH301, and pregelatinized starch at compression force range of 1–20 kN

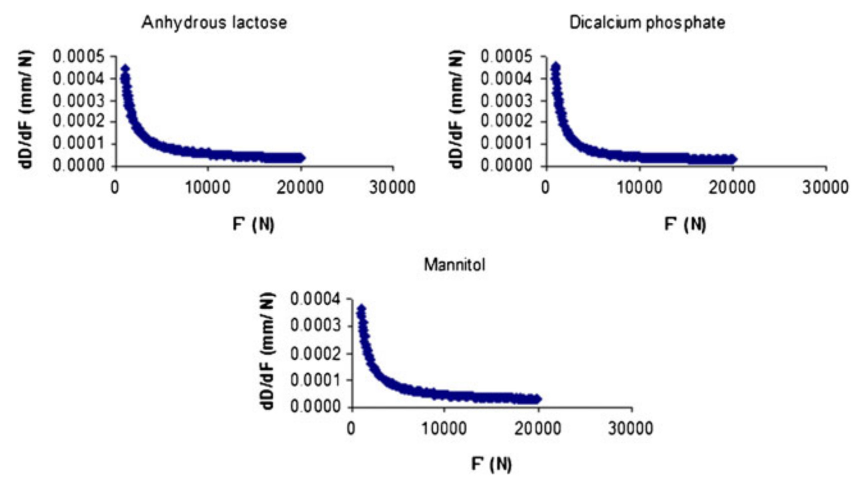

Fig. 3. Relation between first derivative and compression force for mannitol, dicalcium phosphate, and anhydrous lactose at compression force range of 1–20 kN

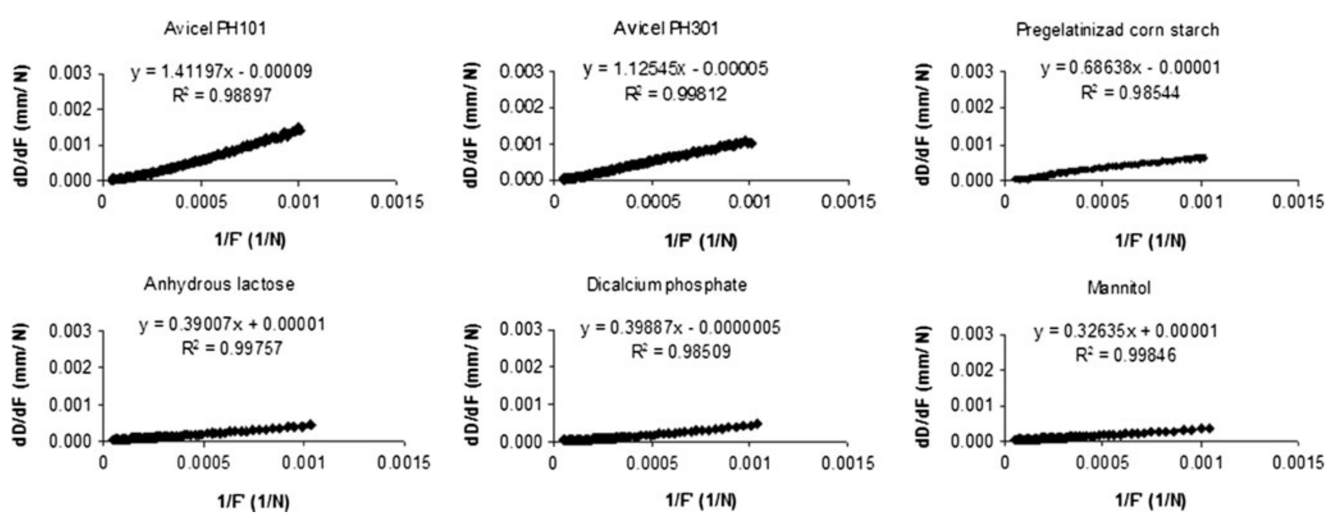

Fig. 4. Representative plots showing the relation between first derivative and 1/F′ for pregelatinized corn starch, Avicel PH301, Avicel PH101, mannitol, dicalcium phosphate, and anhydrous lactose

Table II. Summary of Linear Fit Parameters of the dD/ dF *vs.* 1/ F′ Plots (n=3)

Material	Average slope (mm)	Standard deviation	Average intercept (mm)	Standard deviation	R^2
Avicel PH101	1.42	0.009	0.00009	<0.000001	>0.988
Avicel PH301	1.12	0.004	<0.00001	<0.00001	>0.997
Pregelatinized corn starch	0.69	0.005	0.00001	<0.000001	>0.985
Anhydrous lactose	0.39	0.001	0.00001	<0.000001	>0.997
Dicalcium phosphate	0.39	0.003	<0.00001	<0.000001	>0.983
Mannitol	0.32	0.002	0.00001	<0.000001	>0.998

mannitol), which have slope values of 0.32–0.39. This is consistent with the fact that materials that deform predominantly plastically yield to lower applied pressures when compared to brittle materials. This analysis indicates that the deformation behavior of a compressed powder can be obtained from direct analysis of the displacement *vs.* force profile without the need to know additional information regarding the compressed powder. Due to the limited number of materials studied in this work, the aforementioned needs to be further confirmed.

CONCLUSIONS

In this work, we show how to utilize force *vs* displacement profiles to discern deformation mechanism of powders. Displacement *vs.* force profiles for six powders were collected and analyzed. A linear correlation between dD/dF and 1/F′ is obtained regardless of the deformation mechanism of the powder. Powders that deform predominantly by plastic behavior have larger slope values when compared to those that deform predominantly by brittle fracture. Results from the proposed method are consistent with what's known about the powders. Due to the limited number of materials studied in this work, the aforementioned conclusions need to be further confirmed.

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