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A METHODOLOGY TO CHARACTERIZE
TRIBOELECTRIC CHARGING OF POWDERS

By

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ABSTRACT OF THE THESIS

A Methodology to Characterize Triboelectric Charging of Powders

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The flow of powders and granular materials induces electrostatic charging, and it causes severe problems like agglomeration, adhesions to equipment surface, clogging, separations, and flawed product. Knowledge of charging trends of powders can be effective in decreasing its effects. Unfortunately, due to its complicated nature, charging tendencies of polymeric powders of the exact nature are poorly understood, although largely studied in the literature. Moreover, there is no standard and high precision method available for the characterization and comparison of the electrostatic behavior of powders. In this work, charge gain tendencies of microcrystalline cellulose and acetaminophen in contact with stainless steel are evaluated. The electrostatic charge of powders is first analyzed using a commercially available instrument, GranuCharge™. Inspired by the limitations of GranuCharge™ and considering other methodologies described in the literature, a new setup is developed to measure mass flow rate and charge per unit mass simultaneously. The effect of geometrical parameters and the impact of the

powder flowability on the flow pattern is studied to present a final methodology that gives reproducible charge measurements results with a controlled flow pattern. This methodology is suitable to standardize and can compare the triboelectric charging performance of different materials.

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Chapter 1: Introduction

Powders are profoundly involved in our day-to-day lifestyle because they are used in many major industries, including food, construction, pharmaceuticals, cosmetics, catalytic beds, processing of composite materials, including ceramics and hard metals. The pharmaceutical sector widely uses powders and granular materials [1-3]. Tablets, capsules, and dry powder inhalers (DPIs) contains powders and granular materials as an active pharmaceutical ingredient (API) and excipients [4, 5]. Tablets are used for most solid oral dosages [6], and almost 80 % of pharmaceutical products are tablets and capsules [3].

A good manufacturing process circumvents caking, clogging, dissimilarity in dosage, and other problems[7]. As new drugs are invented, many occasions of failure in formulation occur since the powder behavior is not thoroughly understood[6, 8]. Several factors affect bulk powder behavior, including inherent material properties such as electrostatic charging, frictional forces, cohesive forces, interactions with surrounding particles and surface, and external environmental factors such as humidity and temperature[9, 10]. It is essential to have all the possible information about powder flow behavior, the interaction between the grains, and suitable additives, to expedite process development and decrease costs [7, 11-13]. Electrostatic charging of pharmaceutical powders is the focus of this work.

Pharmaceutical materials are insulators, and many of them are small in particle size, cohesive, irregular in shape, and have low bulk density [14-16]. Hence, they are more likely to gather electrostatic charge while processing and transportation, known as tribocharging of particles. Particle tribocharging can originate from contact between similar materials as well as dissimilar materials. Tribocharging is more complex when there are multiple excipients and API mixtures.

Electrostatic charge in powders can cause severe problems in powder handling, manufacturing process efficiency, and product quality. Some of the undesirable effects of triboelectrification include (i) induce and increase particle-particle adhesion, (ii) increase cohesion and agglomeration, (iii) adverse effect on flowability, (iv) adhesion to surface of the equipment, which results in loss of material and blockage of smaller areas, (v) segregation and poor uniformity (vi) dust explosions. All the above parameters affect process efficiency and product quality [6, 11, 17-25]. In manufacturing capsules, triboelectrification can increase waste as capsules acquire charge while filling process[26]. The tribocharging of particles is a severe problem in the DPIs, as they usually have small size ingredients (1–5 μm APIs with 50 – 200 μm carrier), and particles adhere to walls of the device, which affects the drug delivery [15, 26, 27].

There are some benefits from tribocharging of powders. API-excipient pair with opposite charging nature is the most suitable for formulation as it

can improve the mixture uniformity [28]. Triboelectric separators can be used to segregate two materials with a difference in charging tendency [24]. Electrostatic forces can also be used for powder coating applications [25, 28]. The reduction in size segregation and the coefficient of mixing variation is possible by taking advantage of tribocharging [6].

Several mechanisms can induce electric charge in powders including, impact, rolling, sliding[29]. In the processing of powder, particle-particle interactions and particle-surface of the equipment interactions are both causes of powder tribo-electrification[19]. In high shear processes such as mixing, shear and other forces can induce electrification of powder particles[30]. Usually, pharmaceutical powders have high electrical resistance, which prevents charge dissipation [31]. Charge dissipation also depends on material and environmental conditions[32]. To anticipate the extent, polarity, and magnitude of charging is difficult [33]. Regardless of the difficulties, various physical and chemical characteristics, environmental conditions like relative humidity, as well as time, area, and the surface of contact, are known to affect triboelectrification of bulk powders[6, 18].

Many researchers came up with different designs for the measurement and characterization of powder tribocharging. Different tribocharging measurement studies involving processes such as a pneumatic powder conveying pipe [16], stainless steel blender [34], twin-screw feeder [35] show

that the powder acquires electrostatic charge. At the same time, processing negatively affects the process.

The powder is poured into a Faraday cup in several characterization studies to measure its electrostatic charge [36]. This type of measurement using a Faraday cup and an inclined metal sliding path is presented in numerous studies [29, 37, 38]. Electrostatic charge characterization of the bulk powder is conducted either by shaking [15, 37] or by flowing in a pipe [39-41] or a chute [29, 38, 42].

For characterization of electrostatic charging by sliding the powder, Naik et al. developed a hopper-chute-faraday cup assembly, and Jallo et al. presented a feeder-pipe-faraday cup assembly [40, 42]. Alternatively, Peltonen et al. developed a pipe-faraday cup approach for measuring electrostatic charging as well as the mass flow rate; they encountered adhesion of the powder on the pipe walls, which affected the charge measurement [41]. These methods tend to focus less on the characterization of poorly flowing materials, which usually have a more charging tendency. The methods presented in the literature also have lower precision. The experimental results of electrostatic charging of the bulk powders obtained by Watanabe et al. and Jallo et al. show as much as 50 % and 30 % variability, respectively [37, 40].

Due to having different mechanistic principles, the results for any material are not comparable among the methods. In addition, the results

among the materials themselves may not be comparable for any method used because of the process's sensitivity to the material's flow properties.

The complex nature of powders makes tribocharging challenging to characterize. This study aims to standardize a methodology to measure powder tribocharging with higher precision for materials with different flowability characteristics. The method needs to be able to compare the tribocharging behavior of different pharmaceutical materials.

In this work, a methodology for the characterization of powder electrostatics is developed progressively in Chapters 2, 3, and 4. The pipe-Faraday cup assembly for measuring triboelectric charge is studied by many researchers [39-41], and the technique is commercialized as an electric charge analyzer, GranuCharge™ (Granutools, Belgium). In Chapter 2, electrostatic charge measurements using GranuCharge™ for cohesive and free-flowing materials are conducted. The channel and Faraday cup assembly is adopted from Ireland P.M. [29], Naik, S. et al. [42] and combined with a vibratory feeder, and this system is studied in Chapter 3 and Chapter 4. Chapter 3 explores two designs, including one with a vibratory feeder chute-Faraday cup while the other one also contains a channel. Both chapters focus on determining the stability and improving the precision of the method to measure electrostatic charge. Building on the findings presented in Chapters 2 and 3, the final experimental setup is shown in Chapter 4, with the final methodology stated in Chapter 5.

Chapter 2: Tribocharging Characterization of Powder by Using a Commercially Available Instrument

2.1 Introduction

GranuCharge™ is an instrument commercialized by Granutools, Belgium, to measure and analyze the electrostatic charges induced in powders. GranuCharge™ is helpful to examine the triboelectrification of bulk powder materials. The principal reason for the rise of charge in flowing powders is the charge exchange of powder particles due to collision with each other and with the walls of the flowing device. The equipment presented here measures both these types of collisions as it has a vibratory feeder and has a long flow path. Figure 2.1 shows the main components of GranuCharge™ used in this work. The equipment incorporates a V-shaped vibrational feeder and a non-vibration rotating feeder, Faraday cup, and a ground connection for the operator. The powder flows from the vibratory feeder to the faraday cup through stainless-steel pipes; Figure 2.2 shows the pathway of powder flow. The charge obtained by the powder due to vibrations and flow is measured when the powder falls into the faraday cup. The V-shape of the pipes is selected to incorporate different charging mechanisms of powder triboelectrification such as friction between grains, friction between grains and wall, and the impact of the grains on the wall at the connection between the two pipes [11].

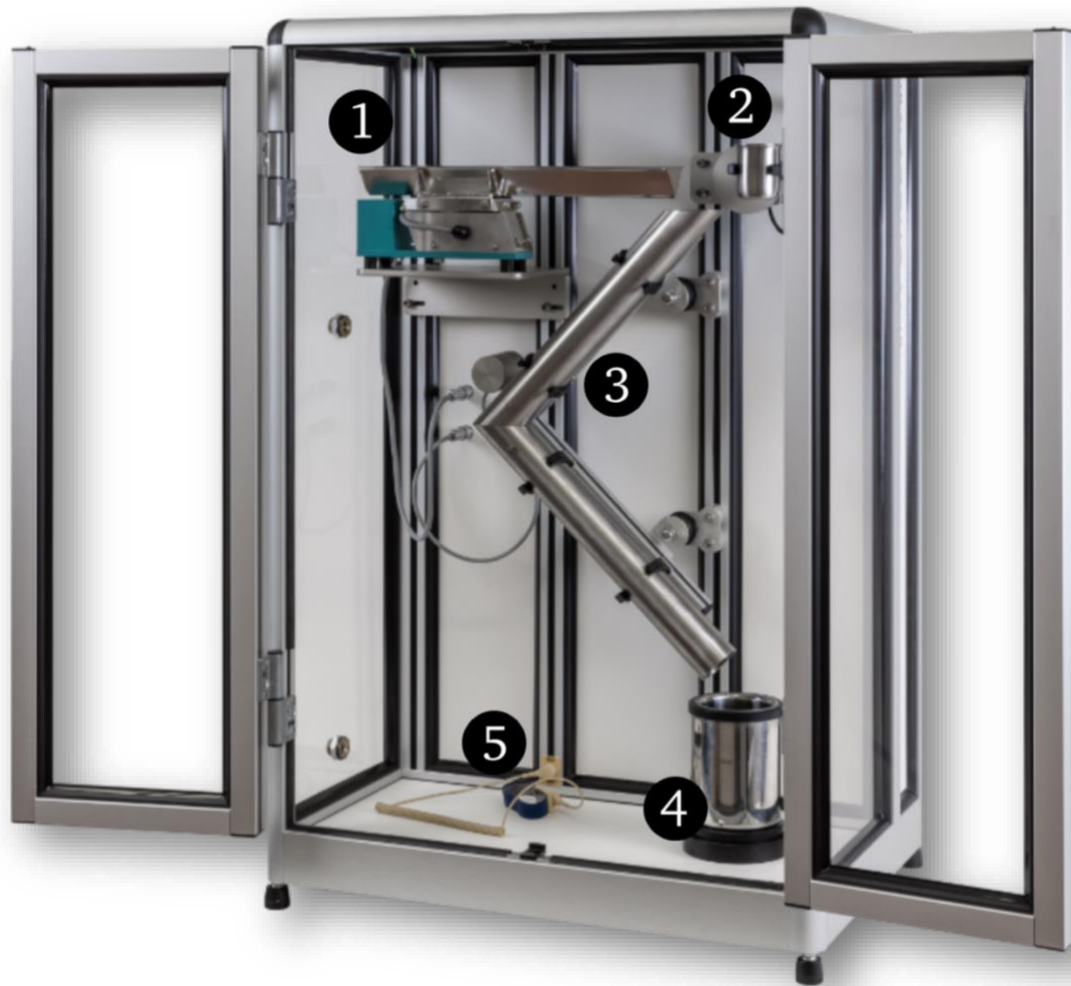


Figure 2.1: Granular material electric charge analyzer, GranuCharge™ (GranuTools™, Belgium); (1) Vibratory feeder (V-shaped chute), (2) Non-vibratory feeder, (3) Stainless-steel pipes (arranged in V-shape), (4) Faraday cup to measure the charge and (5) Ground connection for operator [43].

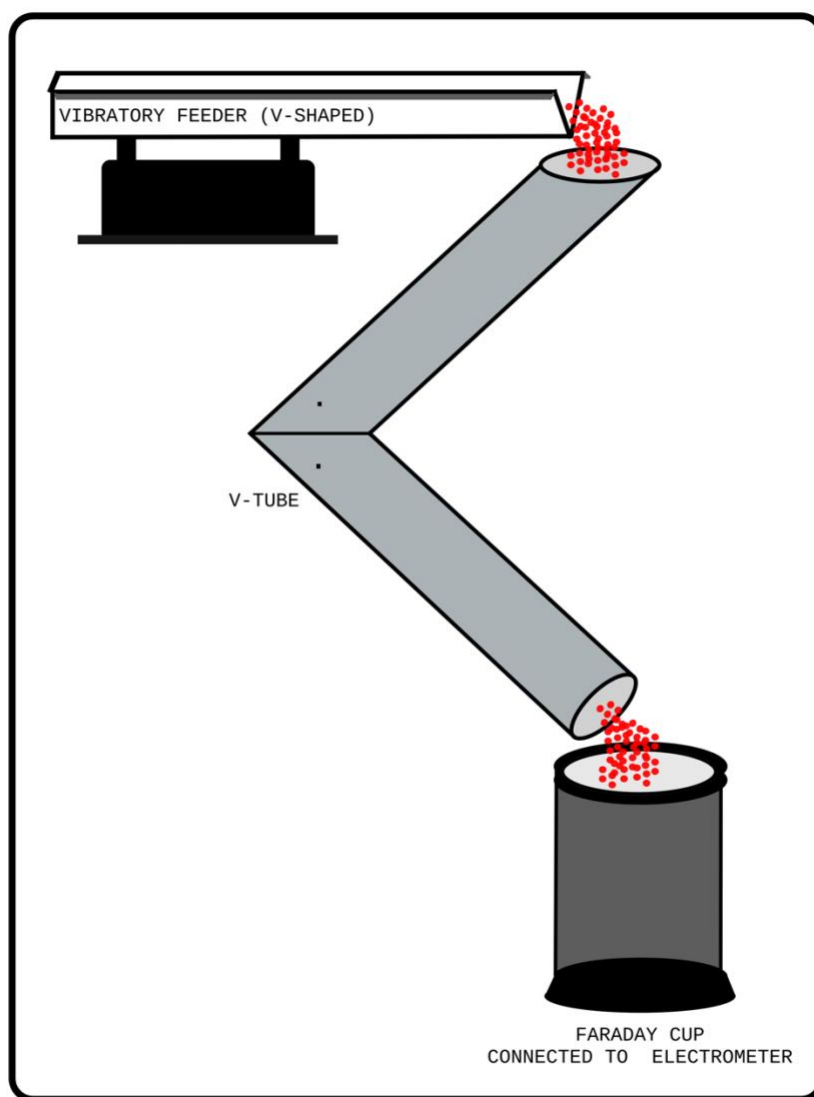


Figure 2.2: Schematic representation of powder flow in GranuCharge™

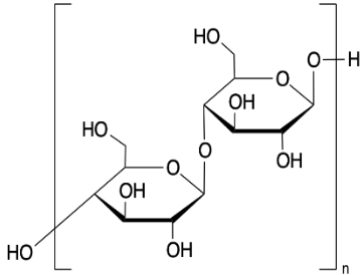
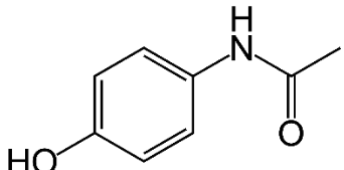
There are several key benefits of GranuCharge™, including (i) the parts such as pipes, feeders, and faraday cup, are easy to clean, (ii) different vibration levels, (iii) relatively fast measurements, (iv) equipment chamber prevents dusting of materials, (v) possibility to measure the initial charge and

(vi) built-in software which has the function for generating graphs, comparison of results, and data export.

2.2 Materials and Methods

GranuCharge™ has 700 mm long (two pipes, 350 mm each) and 47 mm in diameter pipes fixed in a V-shape. The material of the feeder and the pipes is SAE 316 L stainless steel. For the experiments, microcrystalline cellulose (MCC) and acetaminophen (APAP) have been studied. MCC is the most commonly used binder, and APAP is a well-known API [44, 45]. MCC has shown a strong tendency towards adhesion, and APAP is an extremely sensitive API towards tribo-electrification[6, 25]. The experiments utilize different grades of MCC and APAP, such as Avicel® PH-101, Avicel® PH-102, and Avicel® PH-105 from FMC BioPolymer, USA, Acetaminophen Ph Eur semi-fine powder (Semifine APAP), and Acetaminophen Ph Eur powder (Powder APAP) from Mallinckrodt Pharmaceuticals, UK. Table 2.1 shows the properties and molecular structure of the materials. The particle size distribution is determined using Beckman Coulter LS 13 320 dynamic light scattering apparatus, and flow function coefficient (FFC) is measured with an FT4 powder rheometer (Freeman Technology, UK).

Table 2.1: Properties of materials used for Chapter 2

Material		D10	D50	D90	FFC at 6 kPa
 MCC	Avicel® PH-101	24.1	73.5	167.3	6.71
	Avicel® PH-102	37.8	124.7	245.3	13.03
	Avicel® PH-105	6.6	18.6	36.4	2.93
 APAP	Semifine APAP	6.0	23.6	57.5	3.08
	Powder APAP	9.9	57.4	166.0	2.95

2.2.1 The method recommended by GranuTools to measure charge density

To measure powder tribocharging with GranuCharge™, the method recommended by the vendor is followed. The material is dispersed into the Faraday cup manually to measure the initial charge. Subsequently, the final charge was measured using the same material. To calculate the final charge, the material is poured into a stainless-steel vibratory feeder. The material flows from the vibratory feeder into vibratory stainless-steel pipes fixed in a V-shape. During the whole experiment, the pipes are grounded. At the end of the flow, the total charge of the material is measured by the faraday cup. For each

experiment, the amount of material is kept constant at 50 ml (approximated using a glass beaker). The initial weight (m_0) of the material is taken to find initial the charge to mass ratio (charge density $q_0, nC/g$). Final material weight (m_f) is also taken at the end of the experiment. For material adhesion in V-shaped pipes, weight loss ($m_{ad} = m_f - m_0$) is considered. At last, with the help of m_f the final charge density ($q_f, nC/g$) is measured. The charging tendency, which is a charge gained by the material due to vibration and sliding from the pipes, is expressed as $\Delta q (nC/g) = q_f - q_0$. For reused material experiments, the same material is used consecutively to observe the charge density change.

All experiments for one material are performed with the same batch of material, stored at identical conditions. Assessment of initial and final charge is performed at room temperature and room humidity levels. The range of humidity and temperature is 10 – 40% RH and 18°C – 24°C, respectively. During the whole operation of feeding the material into the faraday cup and in the vibratory feeder, the operator is grounded by using the inbuilt bracelet provided with the equipment. The pipes, feeder, and faraday cup are cleaned with compressed air, brush, and paper after each experiment. Between each set of experiments and new material, pipes are washed, the faraday cup and feeder chute are cleaned and wiped with wet paper. They were dried with

Table 2.2: Information about experiment sets and replicates for each material

Material	Experimental Sets	Replicates in each experimental set	Material is reused for replicates
Powder APAP	PAPAP-1, PAPAP-2	3	Yes
Avicel® PH-101	A101-1, A101-2	3	No
Avicel® PH-102	A102-1, A102-2	3	No
Avicel® PH-105	A105-1, A105-2, A105-3	3	No
Semifine APAP	SAPAP-1, SAPAP-2	3	No

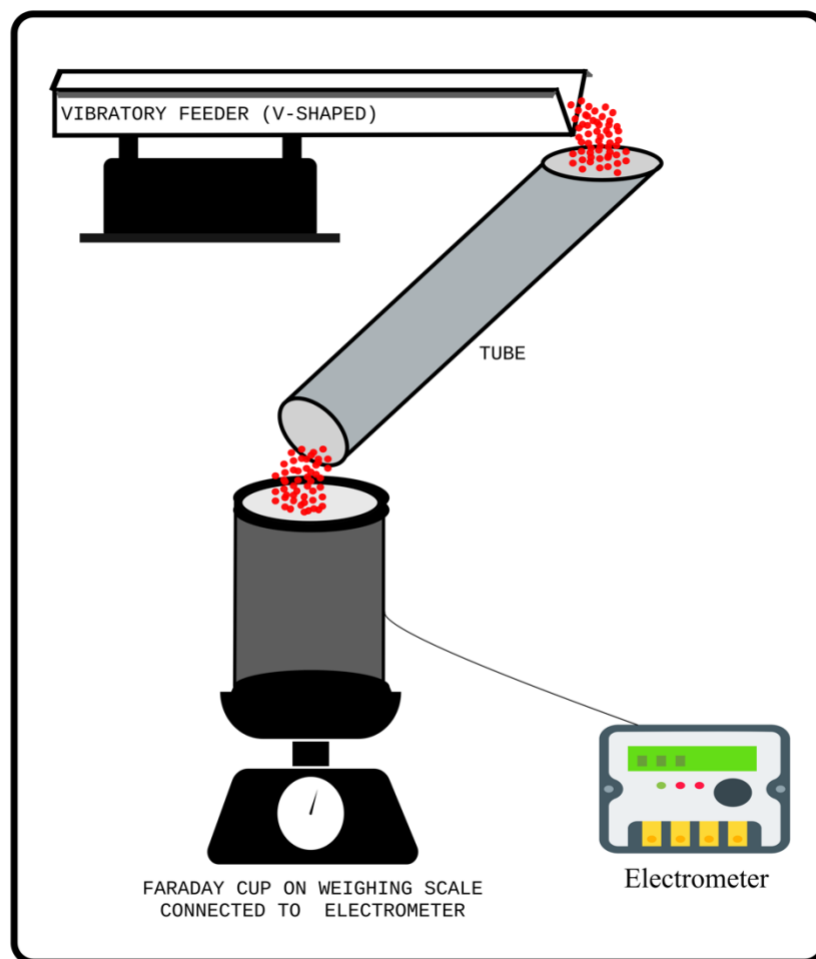


Figure 2.3: Schematic diagram of setup used to calculate mass flow rate and charge density simultaneously using GranuCharge™ equipment

compressed air and kept at ambient conditions for 4 to 12 hours before subsequent experimentation.

Nine sets of experiments are performed to measure charge density. Each experiment set is performed on a different day, with identical conditions. Each experiment set has three replicates performed on the same day with fresh material for every replicate. The error bars indicate one standard deviation calculated with the replicates of the experiments. For powder APAP, two experiment sets are conducted with three replicates in each set. The material is reused for the replicates in powder APAP to find the charge gain change. The sets of the experiment are shown in Table 2.2.

2.2.2 Method to measure the mass flow rate (ad-hoc method)

The total run time is noticed to be different for various runs of a particular material at similar conditions. Therefore it is suspected that the flow rate might be different between runs. To verify that hypothesis set of experiments to measure mass flow rate is conducted. The method used for these experiments is not recommended by GranuTools; it is a make-do method to estimate mass flow rate roughly. One of the pipes from the V-shape assembly is removed, and the faraday cup is placed on a weighing balance. The length of the pipe is 350 mm with a 47 mm diameter. The electrometer measures charge through the faraday cup, and at the same time, the weighing balance measures

the weight. This setup gives mass flow rate and electrostatic charge acquisition values with respect to time simultaneously. The modification is implemented in the GranuCharge™ chamber, and Figure 2.3 shows the diagram of this setup.

2.3 Results and Discussion

2.3.1 Investigation of experimental reproducibility

Two experimental sets are performed for Powder APAP, named experiment set PAPAP-1 and PAPAP-2. The same material is used in three consecutive runs for one experiment set. The motive of doing this is to find maximum charge gain for a particular material and observe charge acquisition tendency when a material is reused. Figure 2.4 represents the charging tendency (Δq) for the experimental set PAPAP-1 and PAPAP-2. The material adhesion in run 1, run 2, and run 3 for PAPAP-1 was found to be 20.5 %, 26.7 %, and 18.2 % of initial weight, respectively, while for PAPAP-2, it was found to be 15.2 %, 18.8 %, and 29.8 % of initial weight respectively. After three runs in both the experimental sets, the total material adhesion was 48 ± 0.3 % of initial weight. In PAPAP-2, a decreasing trend for the absolute charge is observed, which is reasonable because, after material adhesion from the first run, the initial weight is less for the second run and so on. However, due to poor

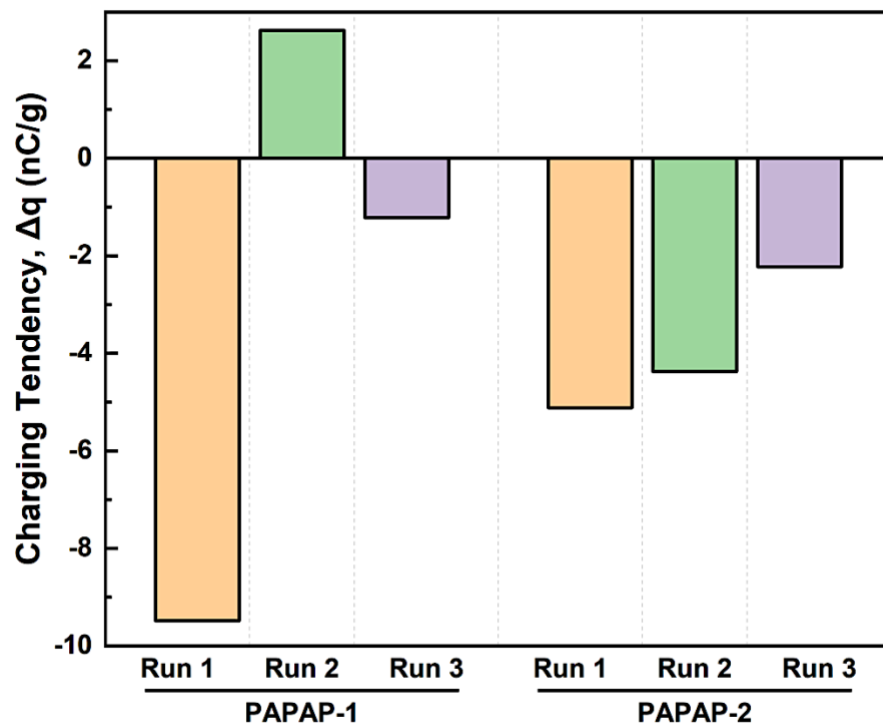


Figure 2.4: Electrostatic charging tendency, Δq in Powder APAP for two experimental sets PAPAP-1 and PAPAP-2, the material is reused in consecutive runs in an experiment set

accuracy, any direct relation could not be deciphered. From this experiment, it is concluded that reusing the material will lead to erroneous results.

Figure 2.5 and Figure 2.6 represent the charge acquisition results for Avicel® PH-101 and Avicel® PH-102, respectively. In Figure 2.5, the results of two sets of experiments are shown, experiment set A101-1 and A101-2. Similarly, in Figure 2.6, results of experimental sets A102-1 and A102-2 are

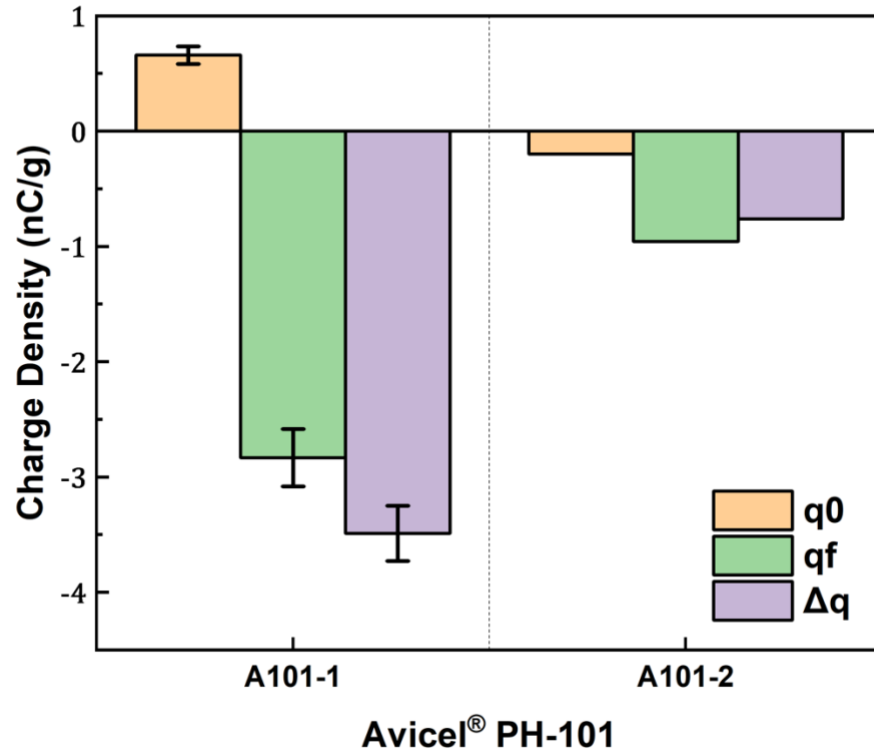


Figure 2.5: Electrostatic charge acquisition for Avicel® PH-101 in the experiment set A101-1 and A101-2. q_0 , q_f and Δq represent initial charge density, final charge density, and charging tendency, respectively

shown. Here, it is noted that all sets of experiments (namely A101-1, A101-2, A102-1, A102-2) are performed on different days.

The initial charge has a different sign for Avicel® PH-101 for different days. The value for Δq on A101-1 is -3.49 ± 0.24 nC/g, while on A101-2 the value is -0.76 nC/g. This large difference in Δq values is a result of the change in values of q_f and q_0 as Δq (nC/g) = $q_f - q_0$. The signs of q_0 , q_f and Δq stay the same on A102-1 and A102-2; however, there is a wide gap between the

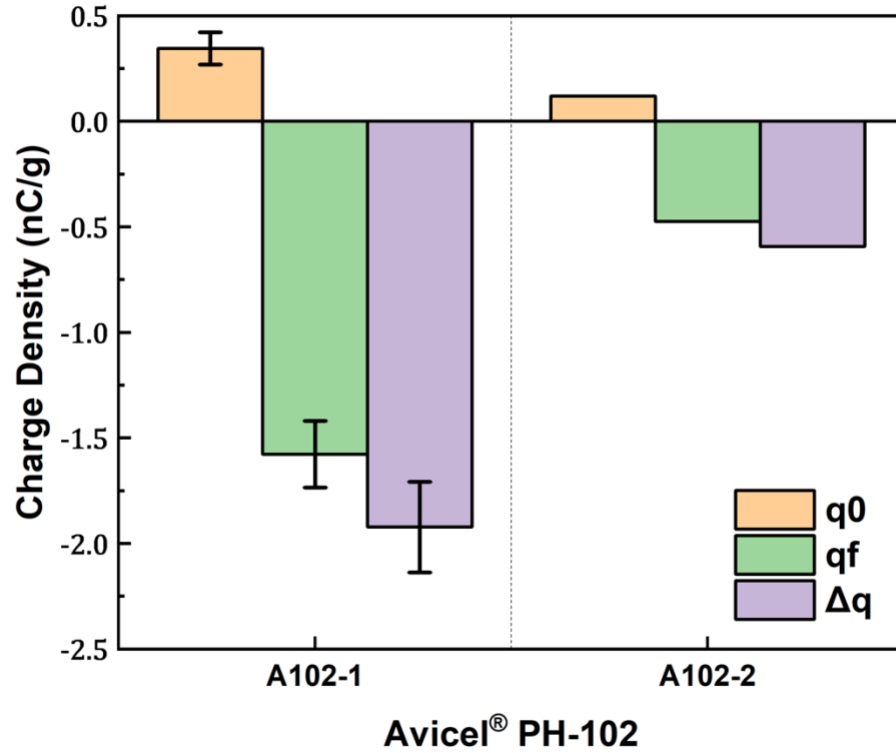


Figure 2.6: Electrostatic charge acquisition for Avicel® PH-102 in the experiment set A102-1 and A102-2. q_0 , q_f and Δq represent initial charge density, final charge density, and charging tendency, respectively

values of q_f and Δq . Material adhesion in the pipes for Avicel® PH-101 and Avicel® PH-102 is observed to be in the range $4 \pm 0.2 \%$ of the initial mass in all experiments. The error in charge measurement for Avicel® PH-101 is analogous to the results reported by Lumay et al. [46].

Similar results for Avicel® PH-105 were found and shown in Figure 2.7. For this material q_0 , q_f and Δq all have different charge signs in different experiment sets. For A105-1, negative q_0 , q_f and Δq (-1.25 ± 0.035 nC/g,

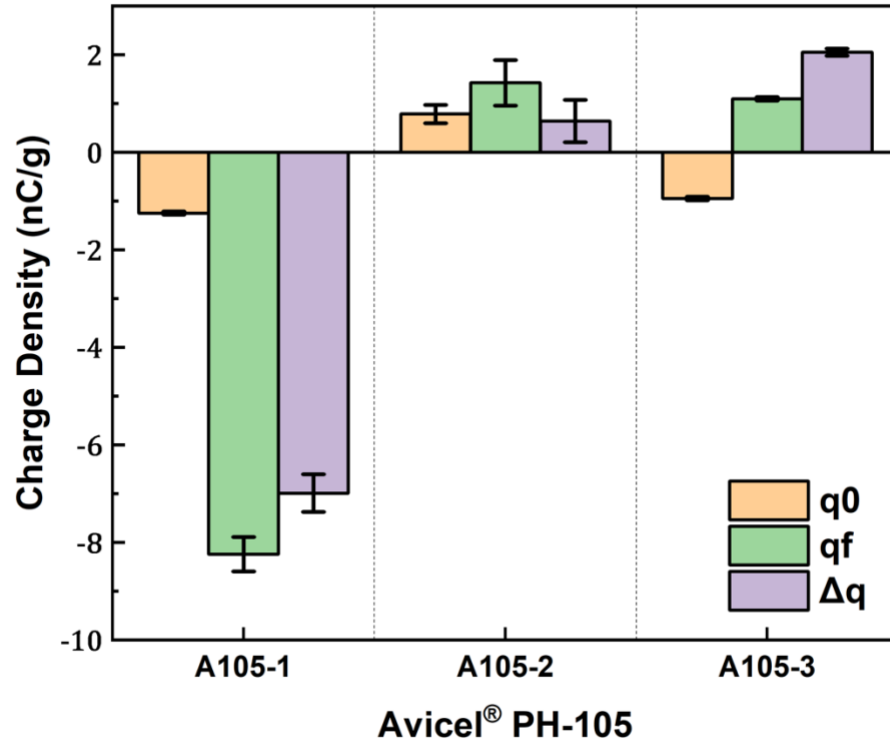


Figure 2.7: Electrostatic charge acquisition for Avicel® PH-105 in the experiment set A105-1, A105-2, and A105-3. q_0 , q_f and Δq represents initial charge density, final charge density, and charging tendency, respectively

– 8.243 ± 0.352 nC/g, – 6.994 ± 0.386 nC/g respectively) are observed, while for A105-2, positive q_0 , q_f and Δq (0.783 ± 0.186 nC/g, 1.423 ± 0.467 nC/g, 0.64 ± 0.434 nC/g respectively) are observed and for A105-3, negative q_0 while positive q_f and Δq (-0.95 ± 0.035 nC/g, 1.095 ± 0.035 nC/g, 2.05 ± 0.071 nC/g respectively) are observed. The material adhesion in pipes for Avicel® PH-105 is found to be 8 ± 3 % of initial mass, which is more than that

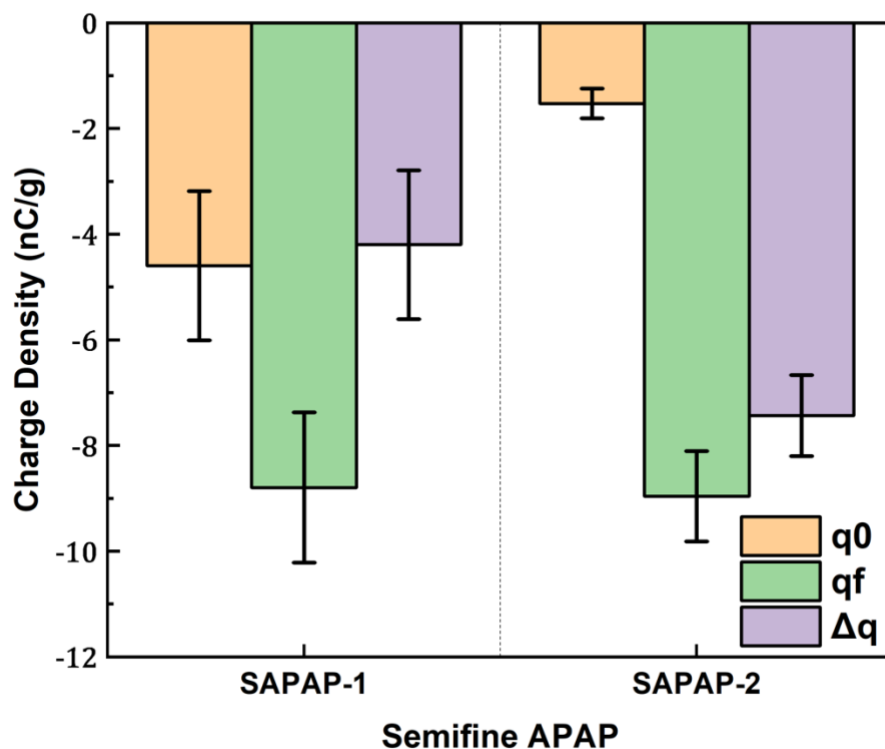


Figure 2.8: Electrostatic charge acquisition for Semifine APAP in the experiment set SAPAP-1 and SAPAP-2. q_0 , q_f and Δq represents initial charge density, final charge density, and charging tendency, respectively

observed in the previous case due to the higher cohesion of Avicel® PH-105, as this is the grade with the smallest particle size.

For Semifine APAP, precision of charge acquisition is studied. The results are analogous with excipient MCC results mentioned previously. Figure 2.8 shows the results for experimental sets SAPAP-1 and SAPAP-2 for

Semifine APAP. The relative humidity is 55 ± 5 % for these sets, the temperature is 20 ± 3 °C, and the initial amount of material is 65 ± 3 ml. The material adhesion for semifine APAP is 17.5 ± 3 % of initial mass, and the error bars represent one standard deviation calculated with at least three replicates of experiments. The sign of charge for semifine APAP is negative for all q_0 , q_f and Δq , but there is a disparity between the values of q_0 and Δq for the two experimental sets. It is observed that the material adhesion is the highest for semifine APAP due to its cohesiveness and its tendency to gain an electrostatic charge, as again, it is the grade with the smallest particle size. The reason for high errors (approximately 40 % for Semifine APAP) can be less control on powder flow. More control on flow is possible by using a straight flow of powder rather than a V-shaped flow. Approximately 30 % variation in measurement of electrostatic charge with APAP for the feeder-pipe-Faraday cup assembly with straight powder flow is observed by Jallo et al. [40].

Table 2.3: Results summary for all experimental sets

Material	D50 (μm)	FFC At 6 kPa	Adhesion (% of initial weight)	Charging Tendency, Δq (nC/g)
Avicel® PH-101	73.5	6.71	4 ± 0.2	-2.13 ± 0.24
Avicel® PH-102	124.7	13.03	4 ± 0.2	-1.26 ± 0.94
Avicel® PH-105	18.6	2.93	8 ± 3	-1.43 ± 4.87
Semifine APAP	23.6	3.08	17.5 ± 3	-5.81 ± 2.29
Powder APAP	57.4	2.95	21.54 ± 5.6	-7.31 ± 3.10

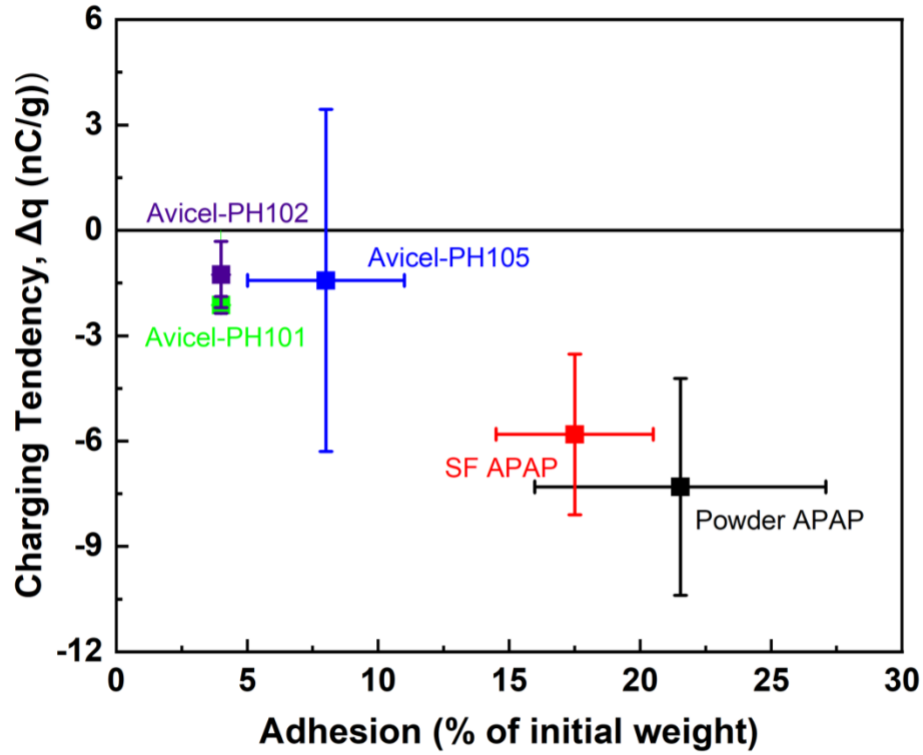


Figure 2.9: Charging tendency, Δq vs. Material adhesion graph for different grades of MCC and acetaminophen

Table 2.3 shows the charging tendency and adhesion values for all materials. Clear relation could not be established for particle size and charging tendency. Figure 2.9 represents the trend of charging tendency with adhesion. Higher adhesion is detected to give higher absolute charging tendency values. While the trend is well established, the data shows very high errors on both axes.

Adhesion of material on pipe walls depends on charge acquisition capacity. Materials with high charging tendency result in higher adhesion. Peltonen et al. observed a significant decrease in charge acquisition after the pipe is saturated with adhesion [41]. Moreover, Murtomaa et al. noticed

electrostatic separation in the pipe while charging measurements [39]. It can be concluded that adhesion is not favorable as it gives erroneous results. Different adhesion patterns are noted for MCC and APAP. MCC particles deposit on the bottom end of the pipe and form a thin layer, while APAP particles thoroughly coat the pipe's walls. It can be expected that at high adhesion, the charge results from more particle-particle interaction and less particle-wall interaction. Moreover, as more material adheres to the pipes, the hindrance to flow increases, and it can cause accidental fall off of the material out of the pipe, which affects the mass flow rate. The adhesion is not controlled; therefore, it will be different at every run, resulting in different charge density values.

The mass flow rate depends on material properties, adhesion of material in pipes, spreading of material on vibratory feeder chute, and intensity of vibrations. The difference in charging tendency can result from variations in the experiment because of the parameters mentioned earlier. To understand this further, the setup of GranuChargeTM was slightly modified, and the results are presented in the next section.

2.3.2 Investigation of mass flow rate

The Faraday cup measures the charge as a function of time. The final weight of the material can be obtained from the material present in the faraday

cup at the end of the experiment, but the instrument cannot measure mass flow rate. The charge acquisition is not constant for the same material in the

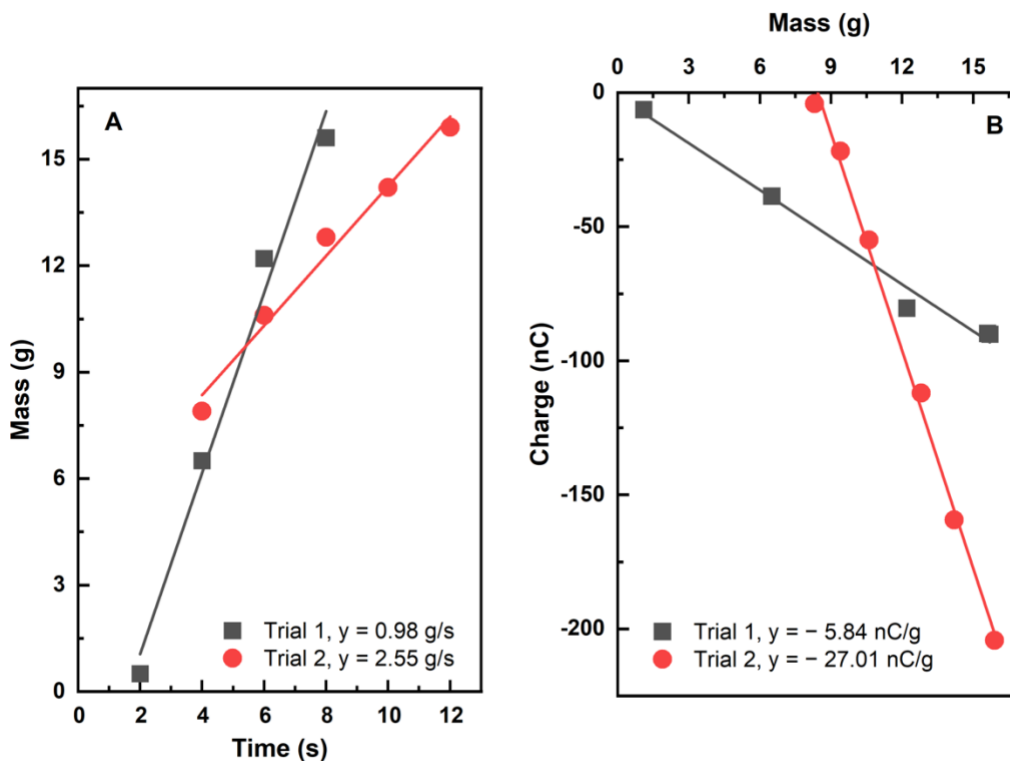


Figure 2.10: Mass flow rate and charge vs. mass for two experiments with Semifine APAP, trial 1 and trial 2, with identical experimental conditions

different experiment set with identical experimental conditions, as shown in Section 2.3.1. To investigate this further, the flow of the material coming out from the V-shaped pipes needs to be measured. The flow of the material in GranuCharge™ is shown in Figure 2.2. To assess the weight of the material and charge acquisition as a function of time, the experimental setup showed in

Figure 2.3 is used. Multiple trials of Semifine APAP are conducted for this setup to observe the mass flow rate and charge as a function of time.

Figure 2.10 shows the results for experimental trial 1 and trial 2 using Semifine APAP. The material adhesion was found to be 4.25 ± 0.6 % of initial weight, which is less than that observed before (17.5 ± 3 %) because of the shorter length of the pipe and the fact that the tube is straight and does not have the elbow in between as seen in Figure 2.3. The observed charge density and mass flow rate are less for trial 1 than for trial 2. Mass flow rate and charge density for trial 1 is 0.98 g/s and -5.84 nC/g, respectively, while for trial 2, the values are 2.55 g/s and -27.01 nC/g. The result shows that the sliding time is different for the same initial amount of material (8 seconds for trial 1 and 12 seconds for trial 2), and the final charge varies drastically.

2.4 Conclusion

GranuCharge™ has a vibratory feeder and long V-shaped pathway for the flow of powders, which incorporates multiple aspects of powder tribocharging. Numerous trials of different materials, including Avicel® PH-101, Avicel® PH-102, Avicel® PH-105, Semifine APAP, and Powder APAP, are presented in this chapter. After several trials of these materials on the same day and on different days, it is concluded that the results are very noisy, especially for highly cohesive materials. High vibration leads to less residence

time of powder on the surface. This decreases the particle-particle and particle-pipe walls contact time. The feeder's size is small, limiting the amount of material used in one experiment. As observed for the materials tested, the initial charge and final charge values vary significantly, not only in amounts but also in the sign of charge (positive or negative). As material adhesion increases, the results have higher instability. It is suspected that a change in mass flow rate due to unexpected adhered chunks flow could be one of the reasons for this variability. After experiments of calculation of mass flow rate, it is found to be not constant. There is a lack of control over the flow conditions. These drawbacks make the results obtained with different materials difficult to be compared and unreliable. To better understand the process, it is necessary to have mass and charge results simultaneously and more control on flow. The experimental setups that can achieve that same are presented in the next chapter.

Chapter 3: Tribocharging Characterization of Powder by Using a Feeder and Channel

3.1 Introduction

GranuCharge™ has some drawbacks, such as the low capacity of feeder, fast time of the experiment, poor accuracy at identical experimental conditions, lack of control on the flow of material as it passes through pipes unavailability to measure mass flow rate, as mentioned in Chapter 2. The need for a robust process that gives mass and charge measurement with time simultaneously is required.

This chapter presents two assemblies (referred to as Setup A and Setup B) using feeder, channel, and faraday cup. Both assemblies (i) are placed in a humidity control chamber, and (ii) can measure charge and mass with time. Essential parameters, namely, the intensity of vibration, angle of feeder chute, angle of the channel, are studied. Setup A and Setup B are identical except for the type of feeder, and the sliding pathway (channel) in Setup B. Figure 3.1 shows Setup A, and Setup B is shown in Figure 3.2. Setup A has no external path for the powder to slide. The powder directly flows from the feeder chute to the faraday cup, while Setup B has a non-vibrating channel after the feeder chute. The powder flows from the vibratory feeder chute and then slides on the surface of the non-vibratory channel to go into the faraday cup.

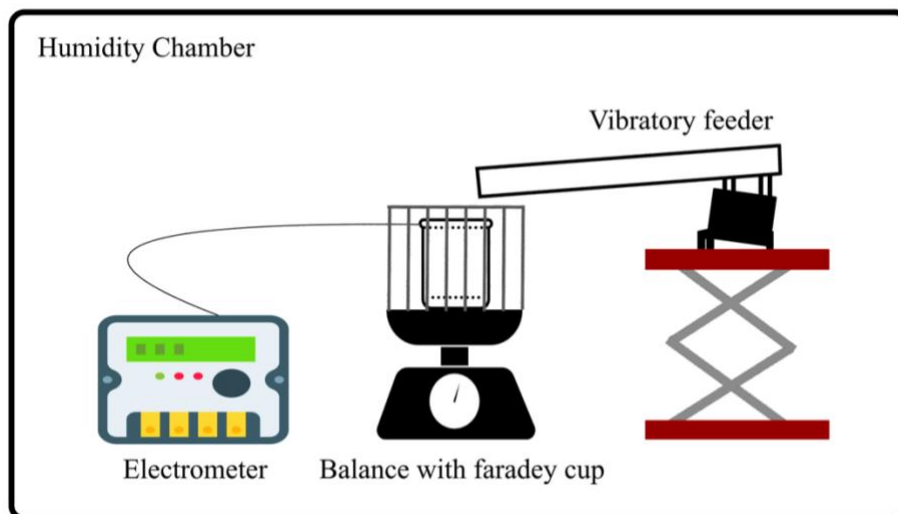


Figure 3.1: Setup A, to measure charge acquisition and mass flow rate using Eriez feeder, faraday cup, weighing balance, and electrometer

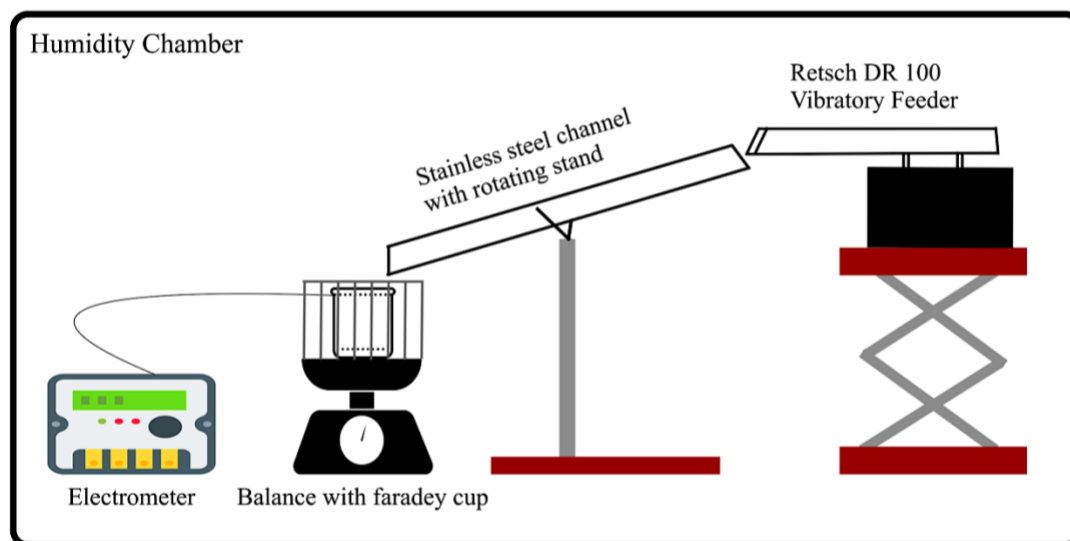


Figure 3.2: Setup B, to measure charge acquisition and mass flow rate using Retsch DR 100 feeder, faraday cup, weighing balance, and electrometer

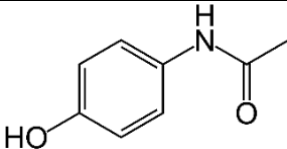
3.2 Materials and Methods

For Setup A, a commercially available vibratory feeder, Model HS from Eriez Manufacturing, Erie, PA, USA, is used. The vibratory feeder has an analog vibration intensity controller, ranging in vibration levels from 1 to 10. The feeder chute's length, width, and height are 500 mm, 106 mm, 65/45 (at closed-end/at the open end) mm, respectively. The Faraday cup comprises two concentric cylinders, the inner one is a cup, and the outer one is a mesh; both are made of aluminum and are separated by a plastic base. The Faraday cup is connected to an electrometer (Keithley 6517A Electrometer / High Resistance Meter, Tektronix, Inc) to measure the charge. To calculate the weight, the faraday cup is placed on a weighing balance (OHAUS Corporation, USA).

The feeder used in Setup B is a commercially available vibratory feeder, DR 100 from Retsch GmbH, Germany. The feeder chute is made with stainless steel (SAE 304) and is 250 in length, 40 mm in width, and 30 mm in height. The channels are 152.4 and 304.8 mm long, 40 mm wide, and 25 mm high. The feeder has a 'standard' and 'extreme' vibration mode with a digital vibration intensity display. For the work presented here, the 'standard' vibration mode is used. The vibration intensity level ranges from 1 to 99 for DR 100 feeder. The polished chute of the feeder and the hot-rolled channel are both stainless steel build.

For the experiments reported in this chapter, Acetaminophen Ph Eur semi-fine powder (Mallinckrodt Pharmaceuticals, UK) and granular acetaminophen (CompapTM L, Mallinckrodt Pharmaceuticals, UK) are used. The properties of materials are listed in Table 2.1. Table 3.1 shows the particle size distribution for CompapTM L. To measure particle size distribution and flow function coefficient (FFC), Beckman Coulter LS 13 320 dynamic light scattering apparatus and FT4 powder rheometer (Freeman Technology, UK) are used.

Table 3.1: Properties of materials used in Chapter 3

Material		D10	D50	D90	FFC at 6 kPa
 APAP	Compap TM L	72.98	307.13	173.25	11.6
	Semifine APAP	6.0	57.5	23.6	3.08

3.2.1 Experimental procedure for Setup A

For the experiment, the desired volume of material is approximated using a glass beaker and kept at a desired relative humidity in an aluminum pan for 48 to 72 hours. The setup is also held in an environment with desired humidity for at least 12 hours before experiment time to ensure equilibrium. During the experiment, the powder is poured manually with a scoop into the

feeder chute. Powder starts to flow on the chute and is dispensed into the faraday cup due to vibration. Faraday cup is connected to the electrometer which measures, and records charge values every 2 seconds. The mass flow rate is calculated via a weighing balance placed under the faraday cup. The feeder and faraday cup chute is cleaned with a brush, paper, and compressed air between each run and washed with water between each set of experiments. After cleaning, it is grounded and left idle for at least 30 minutes to ensure charge dissipation.

The experiments for Setup A are performed using semifine APAP because it has a high charging tendency. All experiments are performed at 10% humidity with a temperature of 22 ± 3 °C and use 100 ml of material. The feeder chute's vibration intensity level and angle are studied to find parameters for the highest charge density and adequate accuracy. The goal is to find optimum values for vibration intensity and angle of the chute to get the highest charge density measurement.

3.2.2 Experimental procedure for Setup B

The experimental conditions for Setup B, such as humidity, temperature, preparation of material, preparation of setup, and cleaning of the chute, channel, and faraday cup, are identical to that of Setup A. Moreover, the working of the electrometer and calculation of mass flow rate is also like Setup A. The total amount of material used for all experiments is 200 ml. The powder is dispensed manually with a scoop on the feeder chute. The flow pathway is

from the feeder (slides due to vibrations) to the channel (slides due to gravity) into the faraday cup. The feeder chute and the channel are grounded while experimenting. The effect on the charge density of three parameters is studied using this setup: (i) the vibration intensity level, (ii) the angle of the non-vibratory channel, and (iii) the length of the non-vibratory channel. To study the parameters (ii) and (iii), the vibration intensity level is fixed at ‘standard’ 60. Two different lengths, 152.4 mm and 304.8 mm, and angular range of 30° to 60° of the non-vibratory channel is studied.

3.3 Results and Discussion

3.3.1 Results for Setup A

It is observed that semifine APAP does not flow at low vibration levels (4 and lower), while at high vibration levels (7 and higher), powder flow is unstable and starts floating in the air. For semifine APAP, different angles from 0° to 30° for feeder are studied. At an angle of 0°, improper powder flow is observed. The angles higher than 15° have high flow rates that result in shorter run times. The faster runs are not beneficial, as it decreases the time of interaction of powder particles with themselves and with the feeder surface. In conclusion, for this feeder, the angle and vibration intensity should be between 5° to 15° and 5 to 6, respectively.

Four sets of experiments are conducted with five replicates in each set named R1 through R5. The objective is to find values of parameters that give high charge acquisition and higher precision. Table 3.2 shows the parameters for four sets and average values of results for five replicates. Figure 3.3 and Figure 3.4 show the graphs for mass flow rate and charge acquisition for the four sets of experiments.

Table 3.2: Experiment parameters and results for Setup A

Experiment	Vibration	Angle of	Mass Flow	Charge
Set	Intensity Level	Feeder	Rate (g/s)	Density (nC/g)
A1	5	10°	0.58 ± 0.17	-2.78 ± 0.61
A2	6	10°	0.67 ± 0.33	-3.17 ± 0.49
A3	5	15°	0.63 ± 0.25	-3.48 ± 0.31
A4	6	15°	0.51 ± 0.11	-3.62 ± 0.30

At a feeder angle of 10°, higher variability in the mass flow rate values and the charge density is observed. The charge acquisition graph for the experiment set A1 and A2 has more variability than that for A3 and A4. The mass flow rates are very unusual for replicates in each experimental set. However, there is no clear trend observed in flow rates as the values are close by and errors are more prominent. Moreover, the value of charge acquisition is almost 20 % higher for experiment set A3 and A4 than A1 and A2. This shows

that for Setup A, an angle of 15° is better than 10° for charge acquisition and accuracy. For mass flow rate, vibration level 6 has better accuracy. While there is a slight difference between results for vibration levels 5 and 6 at an angle of 10° , they can be concluded as indistinguishable at 15° . Results may wrongly give the impression that charge density does not depend on mass flow rate. As shown in Table 3.2, nearly the same mass flow rate is measured for different charge densities. However, as we will discuss in the next paragraph, the flow rate decreases with time for the same experiment, and the charge density also varies.

An increase in thickness of the adhesion layer on the vibrating feeder channel is observed as the experiment progresses. Powders will slide over adhered powder on the channel and, the flow conditions changes during the test. As the angle of the feeder increases, powder adhesion on the feeder chute decreases, and charge acquisition increases. Therefore, for Setup A, a feeder angle 15° and the vibration intensity level 6 are effective for higher charge acquisition. The formation of the adhesion layer and its effects on mass flow rate and charge gain is concerning. Semifine APAP is not a free-flowing but a cohesive material. For free-flowing materials such as CompapTM L or Avicel[®]

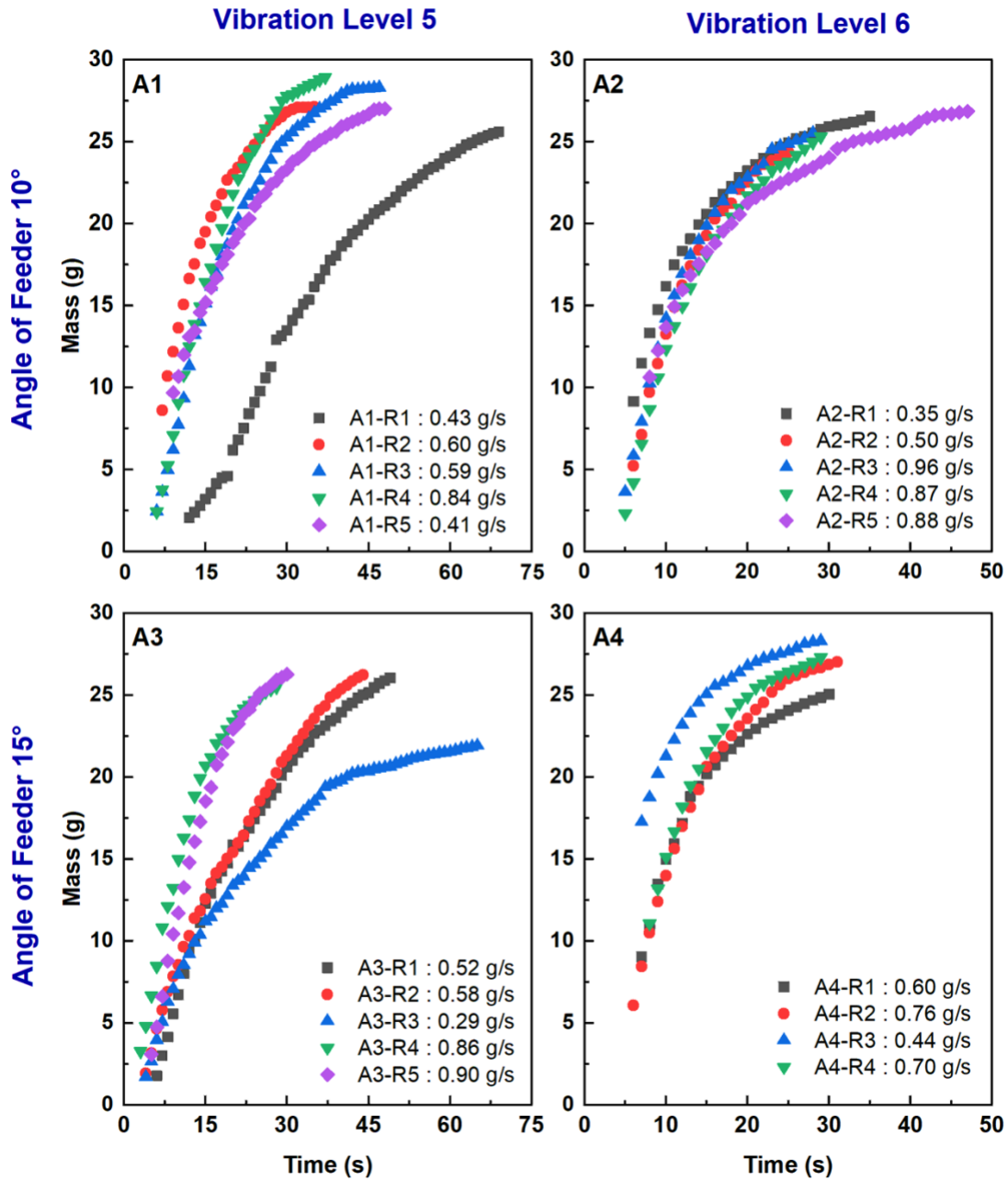


Figure 3.3: Semifine APAP, mass flow rate results of Setup A experiments, four-set, (A1) vibration intensity level 5 and angle of feeder 10°, (A2) vibration intensity level 6 and angle of feeder 10°, (A3) vibration intensity level 5 and angle of feeder 15°, (A4) vibration intensity level 6 and angle of feeder 15° conditions, R1 to R5 shows replicates number for each set of experiment

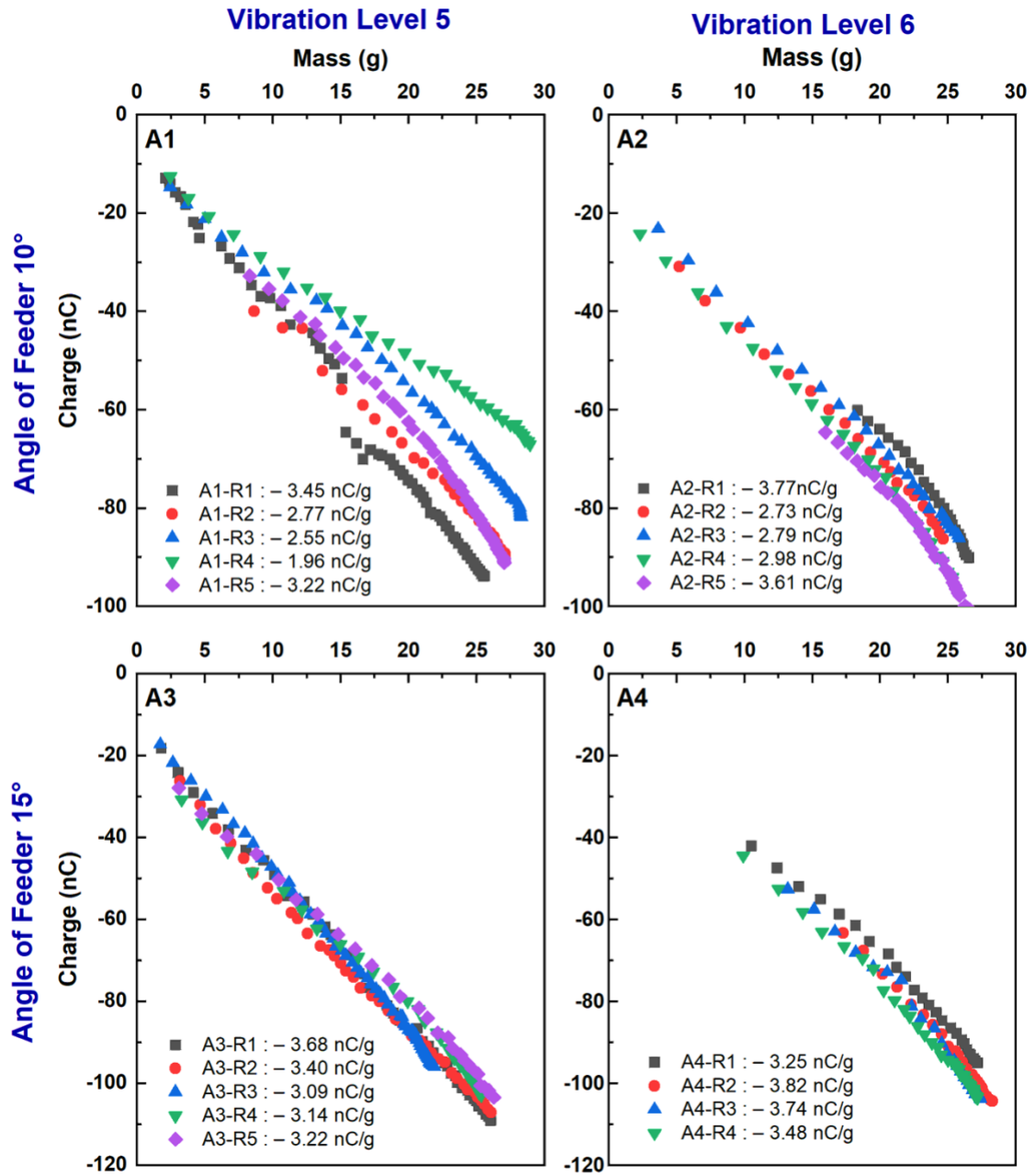


Figure 3.4: Semifine APAP, charge acquisition with respect to the mass of Setup A experiments, four-set, (A1) vibration intensity level 5 and angle of feeder 10°, (A2) vibration intensity level 6 and angle of feeder 10°, (A3) vibration intensity level 5 and angle of feeder 15°, (A4) vibration intensity level 6 and angle of feeder 15° conditions, R1 to R5 shows replicates number for each set of experiment

PH-101, vibration intensity level 6, and angle 15° can be too high. Avicel® PH-101 is found to flow in the air at vibration intensity level 6.

The unsteadiness for the flow rate is observed in all the replicates even with identical parameters, and any relation of it with charge gain could not be established. To compare different kinds of materials, developing a robust method that can work at one set of parameters is imperative. Setup B is designed to achieve this goal.

3.3.2 Results for Setup B

Setup B has DR 100 feeder from Retsch and a channel. During the initial experiments for deciding a suitable vibration range for Compap™ L, it is observed that the powder flow is too slow at vibrations lower than 30, and powder floats into the air at vibrations higher than 60. Therefore, the vibration intensity range for this study is fixed from 30 to 60. Similarly, initial experiments are performed to observe powder flow through the channel at different angles ranging between 10° and 80° . An angle range of 30° to 60° is selected for this study due to similar reasons of either sluggish or high-speed powder flow. Vibration intensity is kept constant at 60 to determine the best values for the channel's length and angle. Four experiments are conducted for the 152.4 mm long channel, and five sets of experiments are performed for the

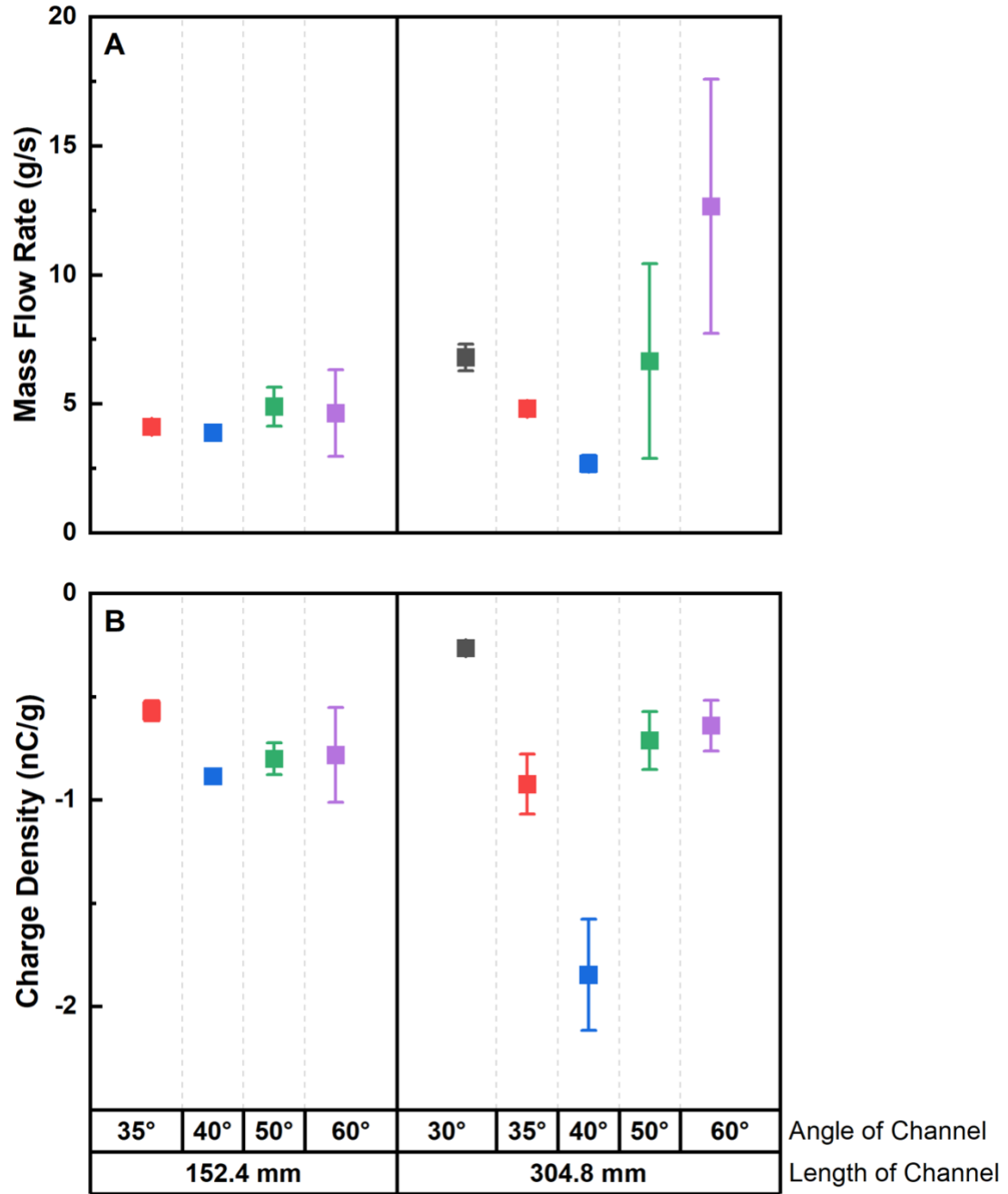


Figure 3.5: Results of variation of angle and length of the channel at fixed vibration intensity level 60 for Setup B, (A) mass flow rate results, and (B) charge density results for Compap™ L

304.8 mm long channel for different channel angles. For Compap™ L, at fixed vibration 60, Figure 3.5 shows the variation of two different channel lengths and five different channel angles on charge density and mass flow rate. The error bar represents one standard deviation of at least 3 experiment replicates for each set. Every replicate in a particular experiment set is performed on the same day.

It is discovered that for the 152.4 mm long channel, mass flow rate, and charge density values are closer to each other, while for the 304.8 mm long channel, values have significant variance. At channel angles higher than 40°, the errors increase. The highest value for charge density is observed for vibration level 60 at a 40° angle of the 304.8 mm long channel. The absolute charge density increases for this channel, and the mass flow rate decreases with increasing the angle from 30° to 40°. This is due to increased powder adhesion on the channel at a high charge density. The powder is observed to form a thin layer on the channel as the experiment progresses. For higher channel angles such as 50° and 60°, the powder flow is more random and fast with shorter contact time between particles and channel surface. Therefore, less charge density and high variation in mass flow are noticed at higher angles. Generally, the error in measurements for Compap™ L increased as the angle of the channel increases; a similar observation for MCC is reported by Naik et al. [42].

To test the reproducibility of results at a channel length of 304.8 mm,

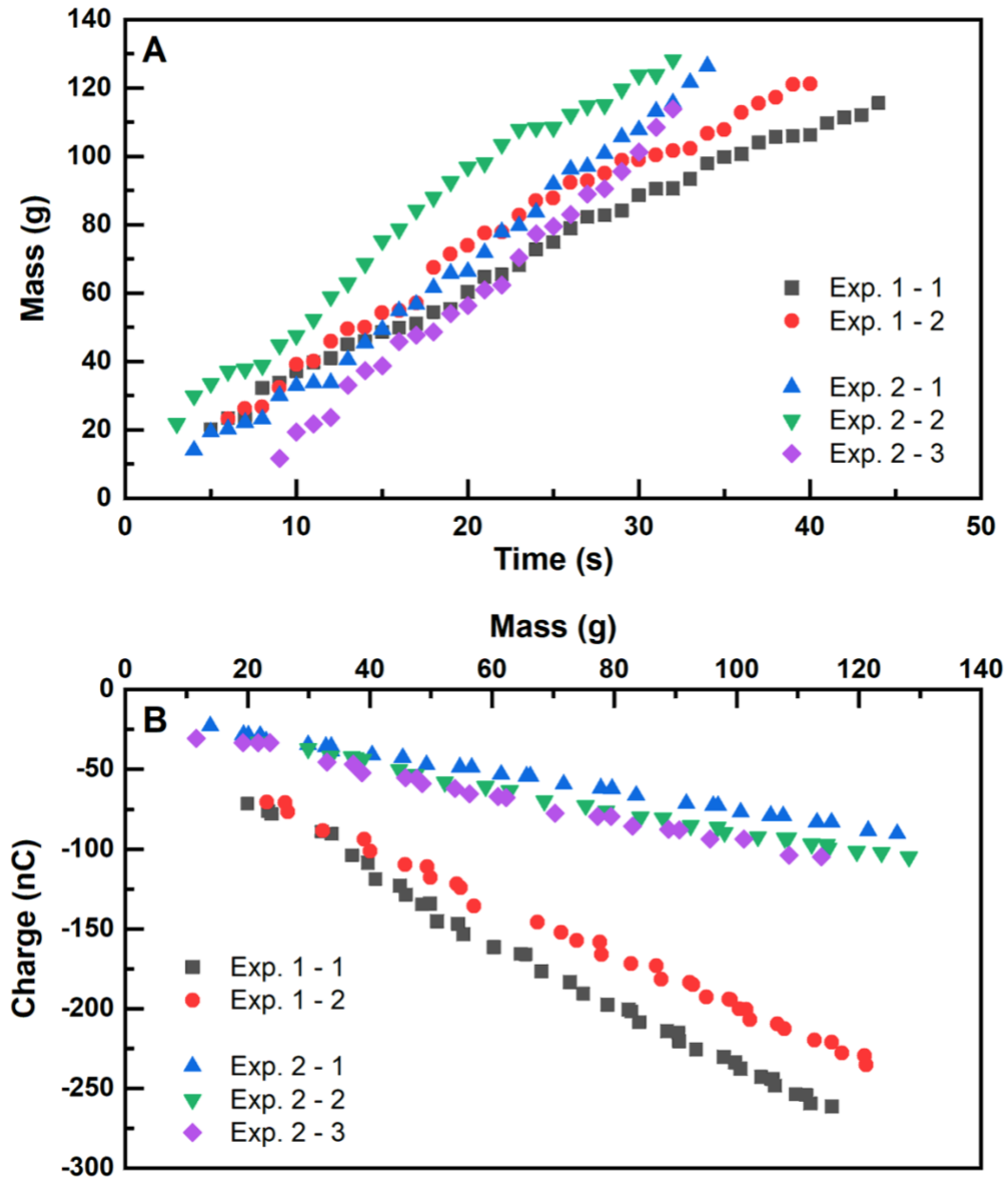


Figure 3.6: Results of Experiment Set 1 and 2 performed on different days under identical conditions, with parameters: vibration level 60, channel length 304.8 mm and channel angle 40° , (A) mass vs. time results and (B) charge vs. mass results, for Compap™ L

channel angle of 40° , and vibration level of 60, two sets of experiments (Exp. 1 and 2) are performed, each on a different day. The temperature, humidity, and sample preparation are the same for both the experiment sets. Figure 3.6 shows the change in (A) mass flow rate and (B) charge with respect to mass. It is noticed that the mass is linear with time, in contrast with what was observed in experiments with Setup A, and it is also true for all results shown in Figure 3.5. The minor variation from linearity in mass flow is observed due to manual scoop feeding of material. From Figure 3.6, it is concluded that Setup B shows poor precision as a decrease of 37 % in charge density is observed for Exp. 2. The flow-through channel is not controlled as it is a non-vibratory pathway. So it is concluded that for the 304.8 mm long channel, results are not reproducible. The reason can be the surface of the channel, which needs further investigation. On account of adhesion on the channel, the flow is variable, and adhered particles can flow as chunks, affecting charge gain. According to the results shown in Figure 3.5 and Figure 3.6, there is no clear trend or relation observed between mass flow rate and charge density for Compap™ L. The results again give the erroneous impression that charge density is not affected by mass flow rate; it is due to the instability of the experiments.

Experiments are conducted to see the effect of cohesive and poorly flowing material such as semifine APAP on Setup B. However, it is observed that semifine APAP does not flow from the non-vibratory channel, as shown in Figure 3.7. There can be two reasons for this, the first being the non-vibrating

nature of the channel and the second being the finishing polish type (hot-rolled in this case) of the channel. To make the surface of the channel smoother, it was brushed with a 1000 grit sandpaper; nonetheless, semifine APAP did not flow.

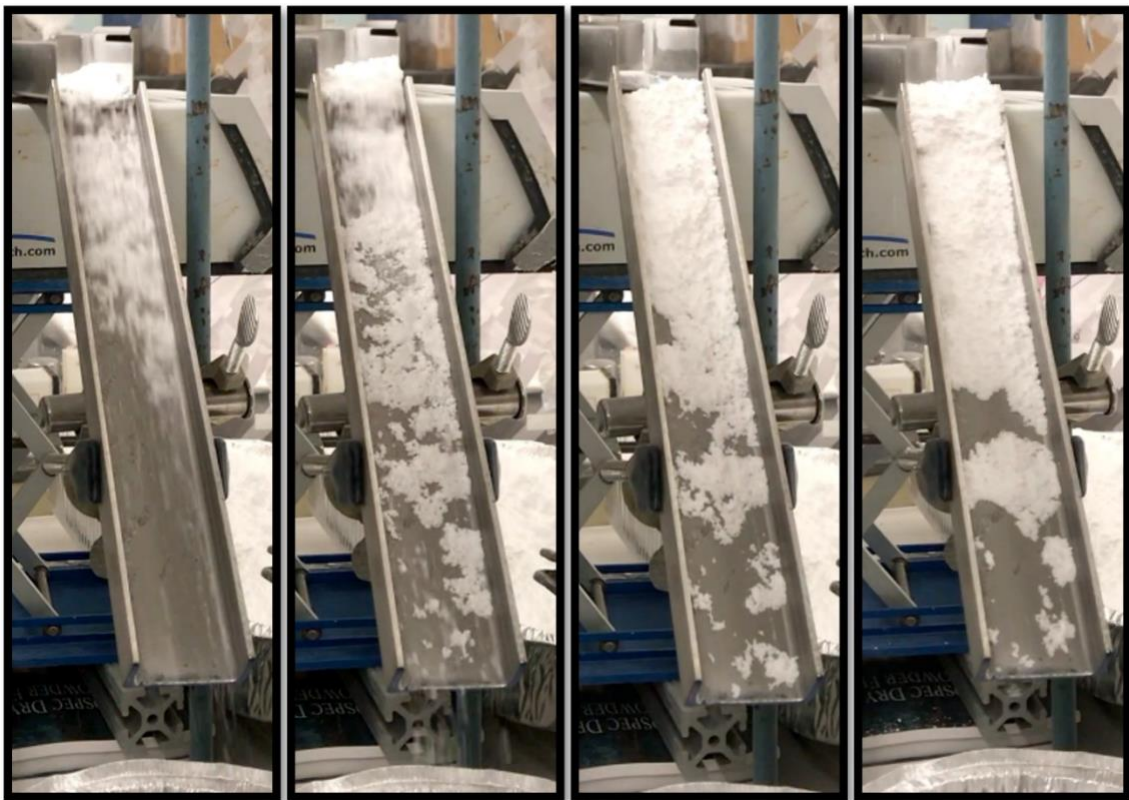


Figure 3.7: Flow of Semifine APAP in the 304.8 mm channel of Setup B at 40° channel angle; as the time of experiment increases, the amount of powder that stuck on the channel increases

3.4 Conclusion

Two setups are presented in this chapter that used different feeders. In Setup A, the powder flows directly from the feeder to the Faraday cup, but in Setup B, powders flow through a channel. The powder gets accumulated on the chute in Setup A and on a channel in Setup B. As the accumulation is random in every run, both setups demonstrate poor accuracy of measurement. The flow rate results show considerable variability. The mass flow rate and charge density values are close to each other at all angles of the channel for channel length 152.4 mm, but they are different for channel length 304.8 mm. Charge gain might be higher when the powder has more distance to travel, with the use of the channel, but flow through the channel is not controlled, especially for cohesive materials such as Semifine APAP. The cohesive and charge-sensitive material gets stuck in the channel or creates perturbation and hinders the flow. The adhesion on the channel increases with the time of the experiment, and it can cause randomized chunks in the flow, which will affect charge gain.

Moreover, experiments conducted on different days provide different results. Material adhesion on the flow pathway also affects the charge density. The thickness of the powder flow layer across length and width should be similar for the whole duration of the experiment to avoid variability. Since it

is not possible with the non-vibrating channel, the final methodology is developed without the channel and present in the next chapter.

Chapter 4: Tribocharging Characterization of Powder at a Controlled Volumetric Flowrate

4.1 Introduction

The two setups shown in Chapter 3 had problems in powder flow-through channels for cohesive materials and precision of data, especially on experiments conducted on different days. There are two working hypotheses; (i) the material, especially cohesive, does not flow very well with gravitational force, and (ii) the manual feeding of material is the leading cause of poor reproducibility. The two main reasons for poor accuracy include (i) the non-uniform distribution of the powder over the chute and (ii) the variation in powder flow layer height due to reason (i) and accumulation on the channel. Random accumulation of powder on the channel leads to the difference in flow between each experiment, resulting in high variability, as seen in Chapter 3. The powder was dispensed on the feeder chute manually, which creates the human error. For each experiment, the weight of the material taken is constant. Still, the distribution of powder over the chute will vary for each run, as it is challenging to have identical distribution over the chute. This issue becomes severe in the case of multiple operators, creating more errors.

Moreover, the powder flow is uncontrolled because the flow layer height will depend on powder distribution over the chute, especially for cohesive materials. As discussed in the previous chapter, it is necessary to have control

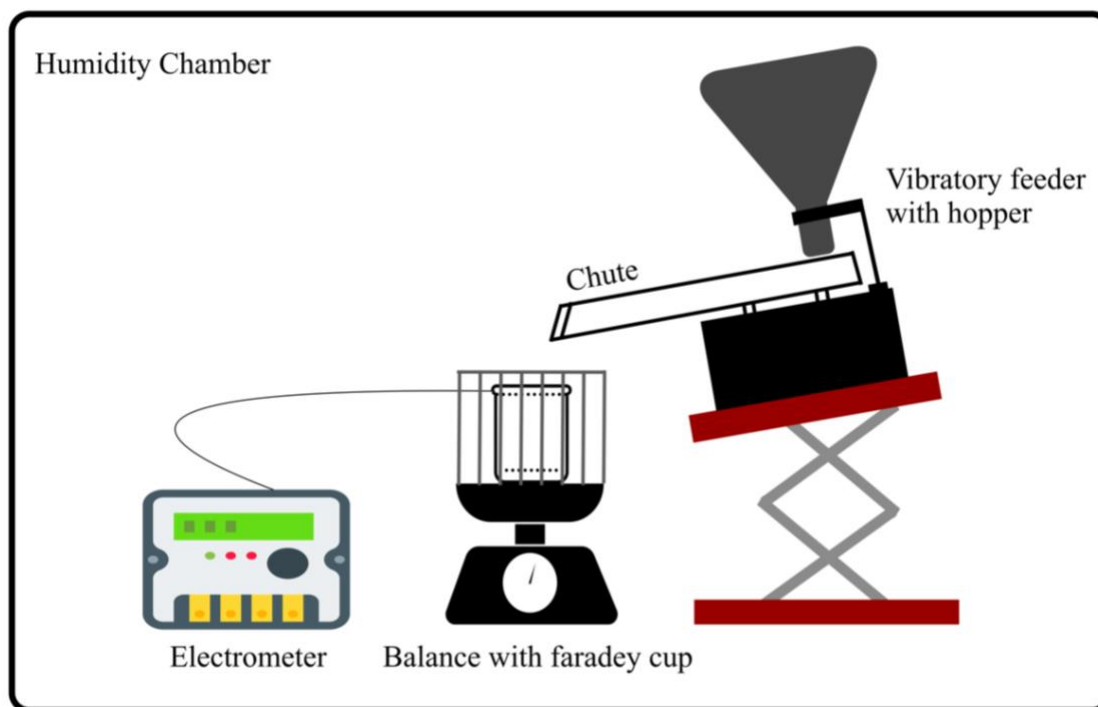


Figure 4.1: Setup C to measure charge acquisition of powders

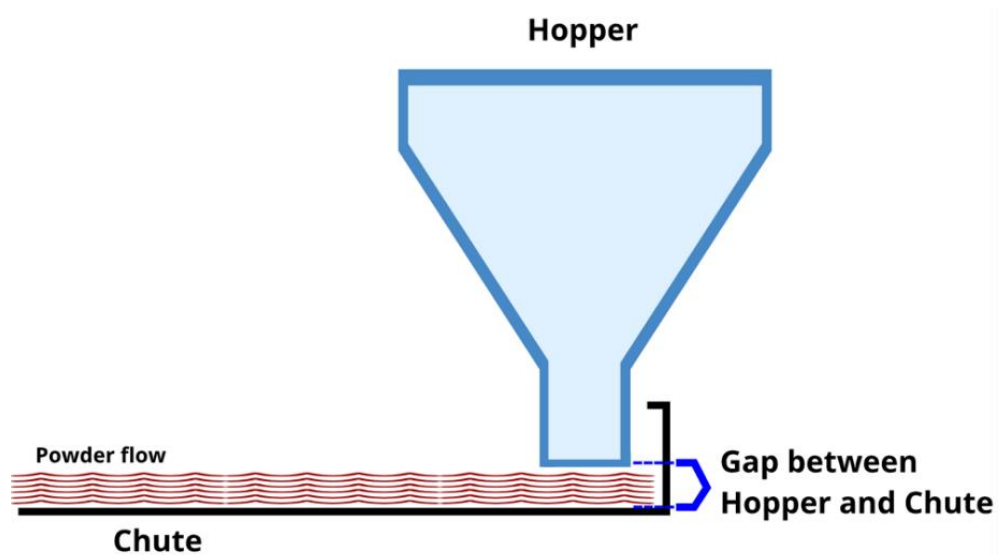


Figure 4.2: Illustration of 'Gap' between hopper and chute

flow layer height and volume. The flow layer volume depends on the flow layer height because the length and width are constant for the chute. To overcome the obstacles mentioned above and control powder distribution and flow layer height, slight modifications are made in Setup B (presented in Chapter 3). The use of the channel is eliminated to make it suitable for all types of materials. Moreover, a hopper is introduced for powder input to have more control on flow layer height and powder distribution. The new Setup C is presented in Figure 4.1.

The hopper eliminates the manual addition of material over the chute and thereby circumvents operator error in powder distribution. The flow layer height is also controlled by varying the distance between the hopper opening and the chute, which can be defined as the ‘Gap’ between the hopper and the chute. The illustration of Gap is shown in Figure 4.2. The introduction of the hopper adds one more parameter to study, the Gap. For this final setup, parameters such as vibrational intensity, angle of inclination of the chute, and Gap are examined. The methodology used to measure charge acquisition for every type of material, in a way that it can be compared with each other, is developed, and presented in this chapter.

4.2 Materials and Methods

The DR 100 vibratory feeder from Retsch GmbH, Germany, is used for Setup C. The stainless steel (SAE 304) chute used in this setup has a constant width and height of 40 mm and 30 mm, respectively, across its length. The length of the chute is 250 mm. The feeder chute is easily replaceable, and Retsch GmbH, Germany, provides the chute. The hopper is made with stainless steel (SAE 304) and has 2.8 liters of capacity with a 30 mm diameter opening. Another chute with length 210 mm, width 75mm, and height 30 mm with a hopper of 3.5 liters of capacity and 60 mm diameter opening is studied to standardize method. The Gap is changed by moving the hopper vertically on the hopper support rod (which is also a scale). The hopper support rod was provided with the feeder. Keithley 6517A Electrometer / High Resistance meter from Tektronix, Inc is used to measure charge gain. Art sand ($D_{50} = 288 \mu\text{m}$) of distinct color is chosen to examine the mixing behavior.

The experiment reported in this chapter, Avicel® PH-101 (FMC BioPolymer, USA), Semifine APAP and Compap™ L (Mallinckrodt Pharmaceuticals, UK), calcium carbonate (Delpharm Pharmaceuticals), and silicified microcrystalline cellulose (Prosolv® SMCC 50 LD, JRS Pharma LP, NY) are used. ProsoLv® SMCC 50 LD combines MCC and colloidal silicon dioxide, making it a versatile excipient. Table 4.1 shows the properties of materials used in this study. Beckman Coulter LS 13 320 dynamic light

scattering apparatus was used for particle size distribution analysis, and FT4 powder rheometer (Freeman Technology, UK) is used to calculate flow function coefficient (FFC).

Art sand of distinct color is placed on the chute for mixing behavior experiments before turning on the feeder. Videos and pictures are taken of the flow to check if there is any mixing due to vibrational flow on the chute. All

Table 4.1: Properties of materials used in Chapter 4

Material	D10	D50	D90	FFC at 6 kPa
Avicel® PH-101	24.1	73.5	167.3	6.71
Semifine APAP	6.0	23.6	57.5	3.08
Compap™ L	72.98	307.13	173.25	11.6
Prosolv® SMCC 50 LD	17.8	62.2	145	10.2
Calcium Carbonate	2.2	9.4	304.8	1.98

experiments are performed with 10 ± 2 % relative humidity and at ambient temperature (22 ± 3 °C). Materials for the experiments are kept in a humidity chamber with 10% humidity for at least 48 hours (about two days) prior to the experiment. For each experimental run, one aluminum pan with the desired amount of material is placed. One-pan per experiment strategy is adopted to decrease movement of material and to reduce errors. The amount of material used for each run is kept constant at 250 ml, approximated with a glass beaker. The Setup C is maintained at 10 % humidity for at least 3 hours before the

experiment to ensure it is equilibrated. The hopper and the chute are grounded during the experiment. All 250 ml of the material is poured into the hopper at once before starting the feeder. Between each experiment, the hopper and the chute are cleaned with a brush, paper, compressed air and kept grounded to reach a neutral charge state for at least five to ten minutes. The powder goes into the faraday cup from the feeder, where an electrometer measures the charge gain, and mass is measured by balance. The electrometer is configured such that it records charge values every one second.

4.3 Results and Discussion

4.3.1 Mixing behavior in the vibrational chute

The results from the experiments for the determination of mixing of material on the chute due to vibration are presented in Figure 4.3. With two and three distinct colors of art sand, no significant mixing is observed. However, a slight drift of material towards one side of the chute is observed (yellow frame highlighted part). Three replicates of the experiments are carried out to confirm the observation. After interchanging the position of the colored sand, more tests are conducted, and identical results are detected. This inclination will likely affect the charge gain; therefore, it is corrected by slightly inclining the feeder laterally.

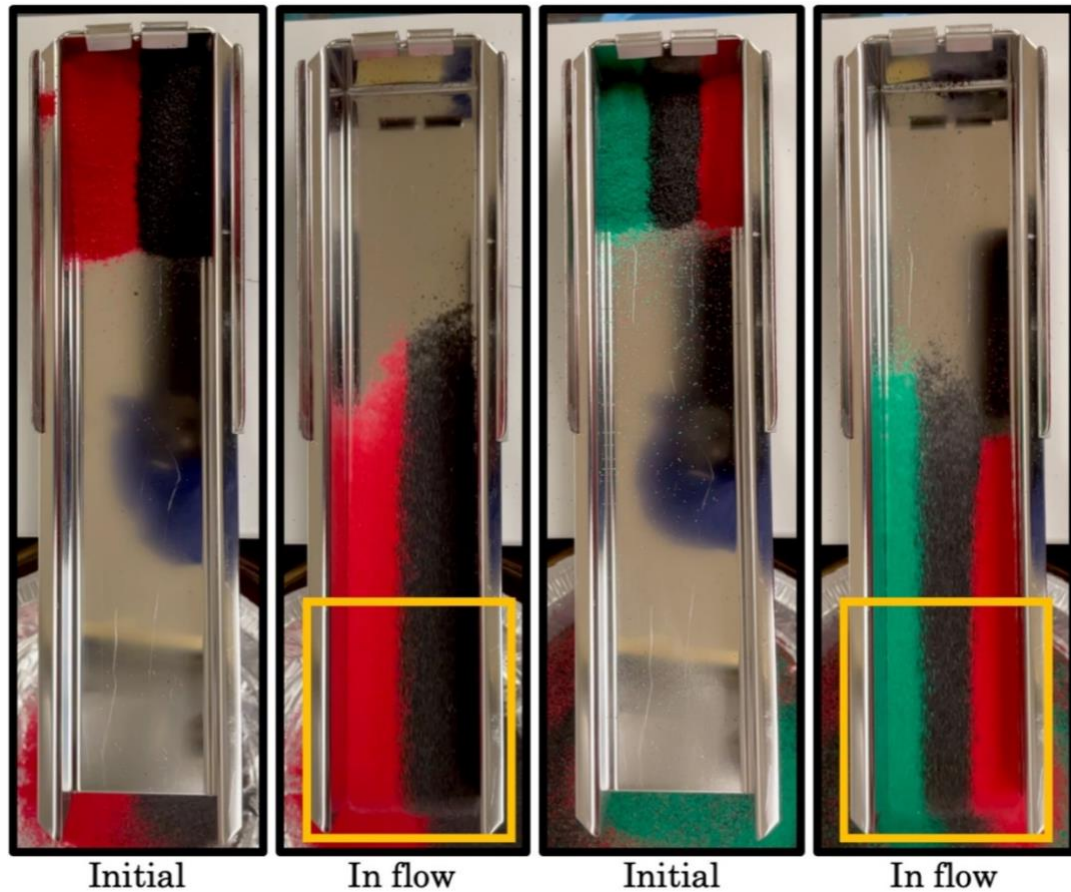


Figure 4.3: Mixing behavior study results with two-color art sand and three-color art sand, the experiment is performed at 40 vibration level, the pictures are screenshotted from video of the flow

4.3.2 Effects of vibrational intensity on charge density and mass flow rate

Variation of vibration intensity from 35 to 60 is explored to understand the effect on charge density and mass flow rate. Figure 4.4 represents the results for three materials. All experiments are performed in triplicates on

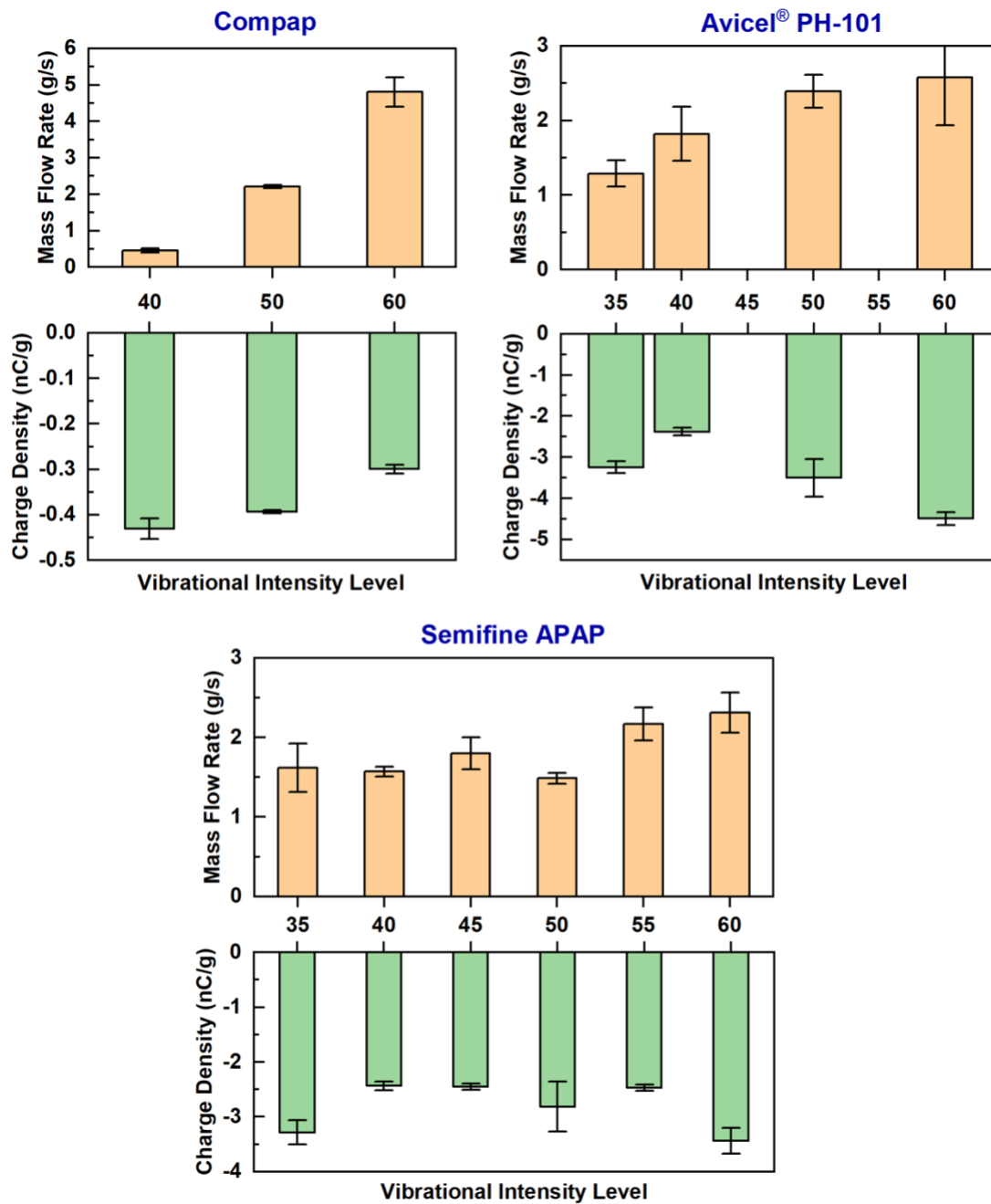


Figure 4.4: Effect of vibration intensity on mass flow rate and charge density for Semifine APAP, Compap™ L, and Avicel® PH-101. The parameters used are chute width of 40 mm, Gap of 8 mm, angle of chute 0°.

different days to ensure reproducibility in measurements. The chute angle and width are 0° and 40 mm, respectively, for all experiments. The powder does not behave well at vibration intensity more than 60, and the flow is very slow at intensities lower than 35. The results for Semifine APAP are obtained with manual feeding as the hopper opening is not wide enough, and the powder gets clogged. For Semifine APAP, similar mass flow rate and charge density are observed for vibration levels of 35 to 50 and 40 to 55, respectively. The manual feeding can be a reason for the similarity of the results, which again proves the importance of the hopper.

Compap™ L and Avicel® PH-101 results are obtained using the hopper with an 8 mm Gap. For Compap™ L, charge density decreases with vibration intensity, while for Avicel® PH-101, it increases. This indicates that charge variation is different for different materials. The reason for this can be the residence time of material over the chute. While the flow rates for both Compap™ L and Avicel® PH-101 increase with vibration, the overall flow rate of the former is higher than the latter. Therefore at a vibration intensity of 60, the residence time of Compap™ L on the chute is much less than that of Avicel® PH-101, which means fewer particle-particle and particle-metal interactions.

Moreover, at higher vibration intensities, the particulate motion will be more intense (increased in the number and/or intensity of collisions between particles), resulting in loss or gain of the charge. The effect of this can be seen in the results for Avicel® PH-101, as the flow rate shows a slight increase from

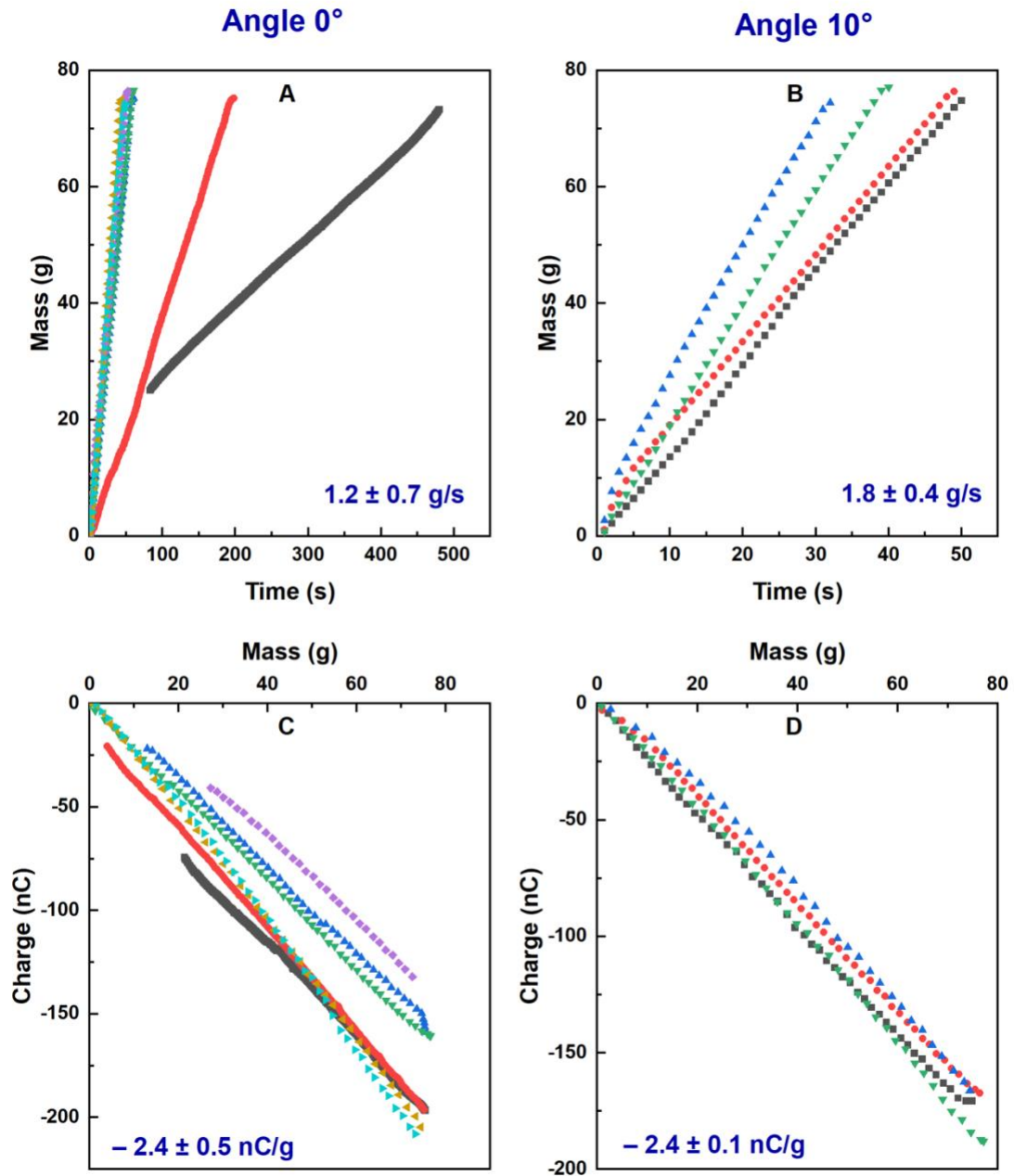


Figure 4.5: Effect of inclination of the chute on charge density and mass flow rate for Avicel® PH-101. (A) mass flow rate at angle 0°, (B) mass flow rate at angle 10°, (C) charge vs. mass at angle 0°, and (D) charge vs. mass at angle 10°. All experiments are performed with vibration intensity 40, 250 mm chute length, 40mm chute width, and 8 mm Gap.

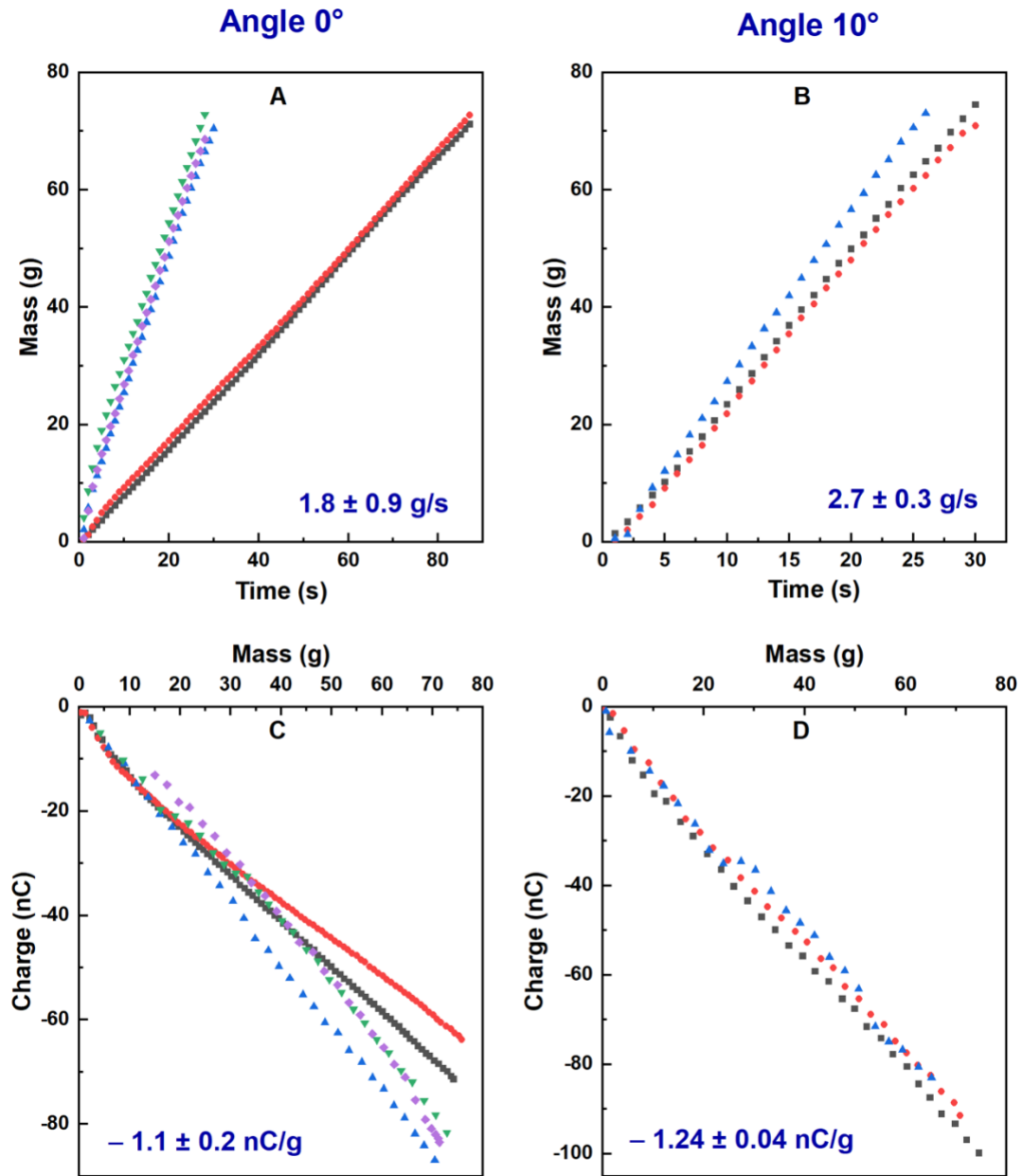


Figure 4.6: Effect of inclination of the chute on charge density and mass flow rate for Prosolv® SMCC 50 LD. (A) mass flow rate at angle 0°, (B) mass flow rate at angle 10°, (C) charge vs. mass at angle 0°, and (D) charge vs. mass at angle 10°. All experiments are performed with vibration intensity 40, 250 mm chute length, 40mm chute width, and 8 mm Gap.

vibration intensity of 50 to 60, but the charge density is increased by 25 %. In general, at vibration intensities higher than 50, more errors in flow are observed. For further experiments, the vibration intensity is fixed at 40.

4.3.3 Effect of inclination of the chute on charge density and mass flow rate

The effect of 0° and 10° chute inclination on mass flow rate and charge gain is studied for Avicel® PH-101 and Prosolv® SMCC 50 LD. The results are shown in Figure 4.5 and Figure 4.6. Replicates of each experimental run are carried out on different days. As seen in the results, large variation is observed at 0° chute angle for both materials on other days. For the same amount of Avicel® PH-101, the total experiment time varies from 50 seconds to 500 seconds (about eight and a half minutes) on different days. A similar variation in experiment time (from 30 seconds to 90 seconds) is observed for Prosolv® SMCC 50 LD. At a chute angle of 10°, the flow is more stable and controlled. The charge density is similar, if not higher, at a 10° angle. For Avicel® PH-101, charge density is the same (– 2.4 nC/g) for both angles. Prosolv® SMCC 50 LD shows more than 10 % higher charge density at 10° chute angle than at 0°. It is concluded that both materials exhibit higher precision and fewer errors at a chute angle of 10°; therefore, for further experiments, the chute is kept at an angle of 10°.

4.3.4 Comparison of charge density for different material

Table 4.2 shows the charge acquisition results for three different materials. The experiments are performed in triplicates and at fixed vibration intensity level 40, Gap of 8 mm, 40mm chute width, and 10° angle of the chute. The charge gain can change significantly if the length is further increased. All the parameters are constant for all experiments, but the powder flow layer volume is not the same due to differences in material densities. Therefore, the comparison between materials may not be relevant. To have an appropriate comparison between materials, the volume of the powder flow layer should be constant for all materials. The Gap can be adjusted for each material to achieve a constant volumetric flow rate. Therefore, the effect of Gap is studied next.

Table 4.2: Charge density results for different material, at vibration intensity 40, the width of chute 40 mm, angle of chute 10°, and Gap of 8 mm (Gap is only for Avicel® PH-101 and Prosolve® SMCC 50 LD)

Charge Density (nC/g)		
Semifine APAP	Avicel® PH-101	Prosolv® SMCC 50 LD
$- 2.40 \pm 0.10$	$- 2.38 \pm 0.09$	$- 1.24 \pm 0.04$

4.3.5 Effect of Gap on charge gain and mass flow rate

The Gap can be varied from 0 mm to 25 mm, as the chute wall height is 30 mm. Figure 4.7 represents the effect of Gap on charge density and mass flow rate for Avicel® PH-101. The parameters for the experiments are kept constant with vibration intensity at 40, the width of chute 40 mm, and chute angle at 10°. At least three replicates for each experiment are conducted on different days to examine data reproducibility. An increase in charge density is detected as flow rates decrease. Charge density is nearly identical for mass flow rates 5.5 g/s to 11 g/s. The maximum error in charge density is observed for Gap 3 mm.

Material holdup can be defined as mass of the powder present on the chute at any point of time. Holdup can be easily calculated with the help of weighing balance. As shown in Figure 4.8, The holdup increases roughly linearly for Gap from 3 mm to 20 mm for Avicel® PH-101, which suggests nearly constant powder bed density. For comparing the charge density of different materials, the relation of holdup vs. Gap can be established by calculating holdup at different Gaps, with vibration intensity at 40 and chute angle at 10°. From the holdup and density of the material, the approximated volume of the flow layer can be found for each material at a specific Gap. By fixing the desired volume of powder flow layer for all materials, Gap and holdup values can be found for each material at that fixed volume. Significant

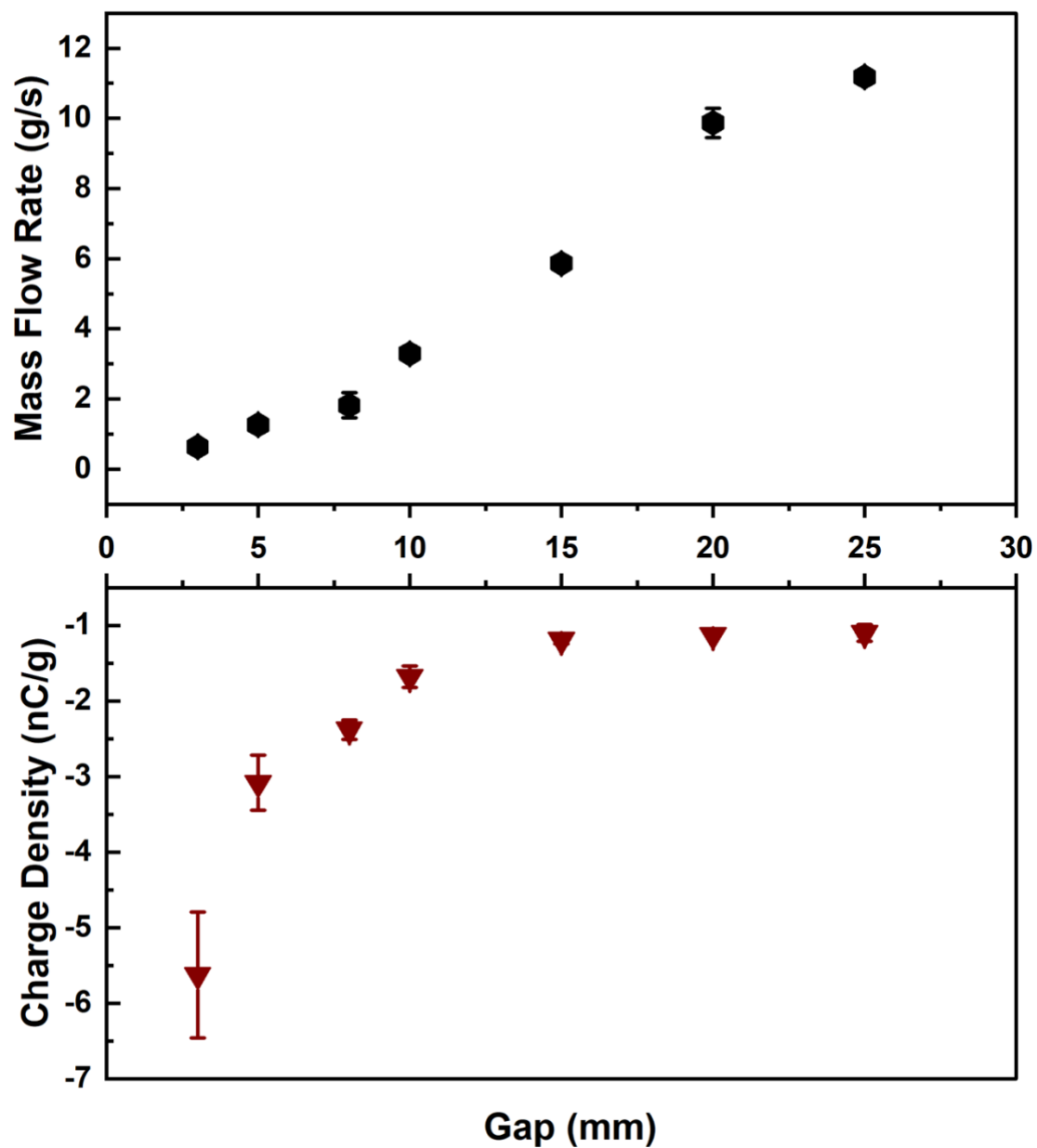


Figure 4.7: Effect of Gap on mass flow rate and charge density for Avicel® PH-101.

The Gap is varied from 3 mm to 25 mm with vibration intensity at 40 chute width 40 mm and a chute angle at 10°.

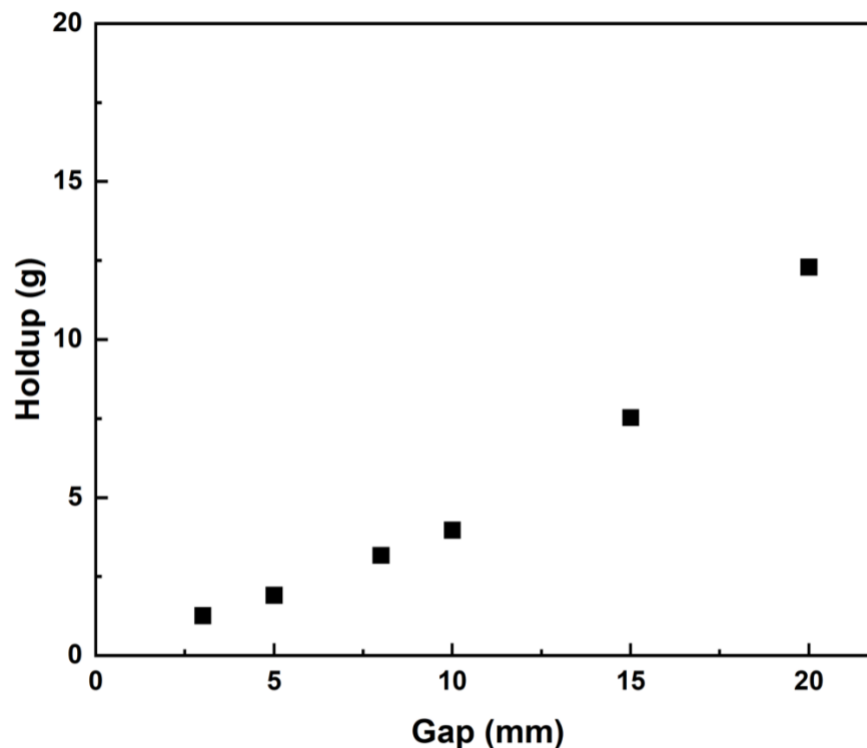


Figure 4.8: Holdup vs. Gap for Avicel® PH-101 with vibration intensity at 40 and chute angle at 10°.

comparison of charge density between various materials can be achieved by doing experiments at those Gap values.

It is observed that cohesive materials such as Semifine APAP are clogged in the hopper with a 30 mm opening diameter. To standardize the method, measuring charge for a wide range of material flowability is necessary. For this purpose, a bigger hopper with a 60 mm opening diameter and 75 mm wide chute with 210 mm length are used. The relation between holdup and Gap is studied for five materials, namely, Avicel® PH-101, Semifine APAP,

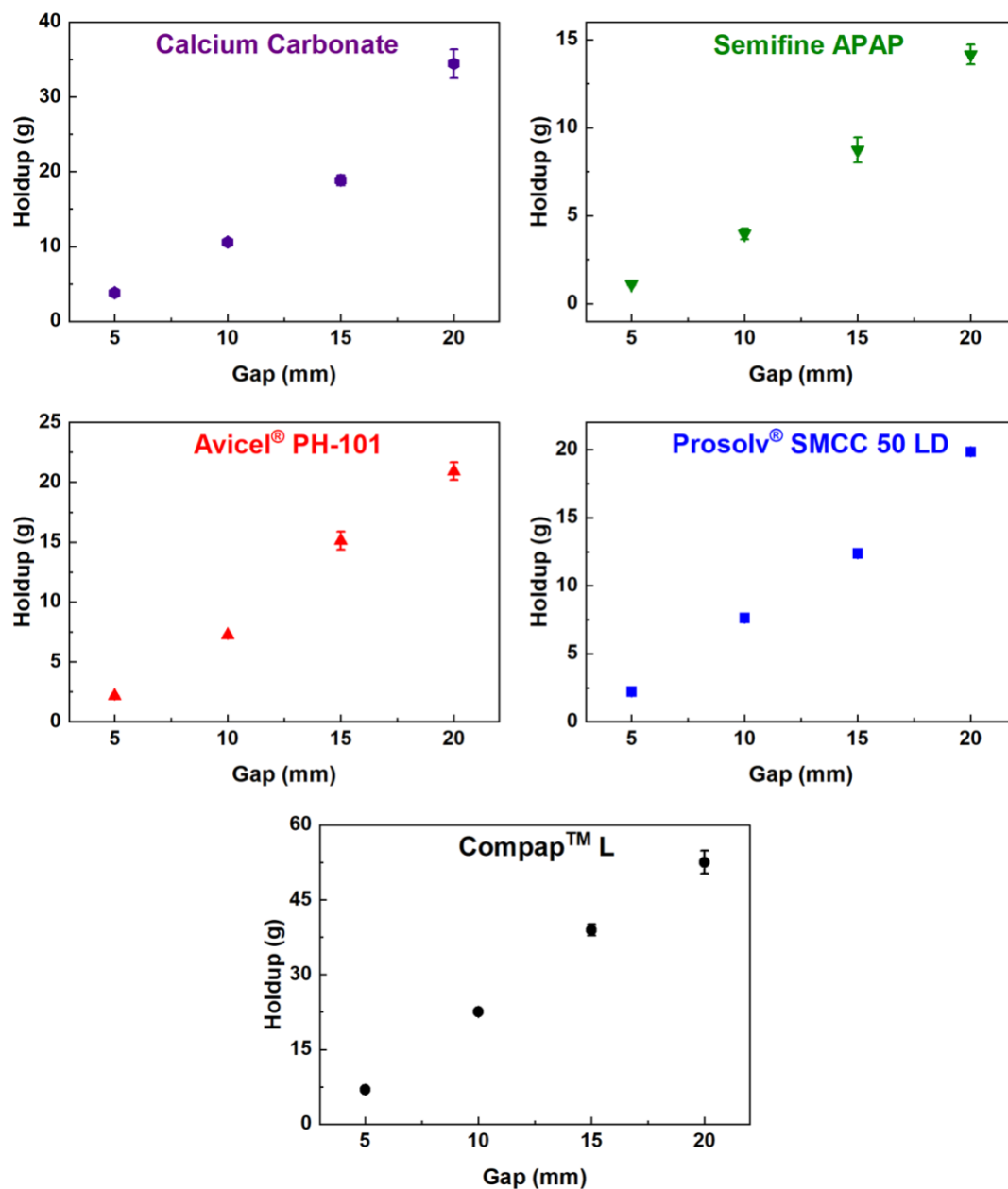


Figure 4.9: Holdup vs. Gap for Calcium Carbonate, Semifine APAP, Avicel® PH-101, Prosolv® SMCC 50 LD, and Compap™ L at chute width 75 mm, chute angle 10°, chute length 210 mm, and vibrational intensity 40.

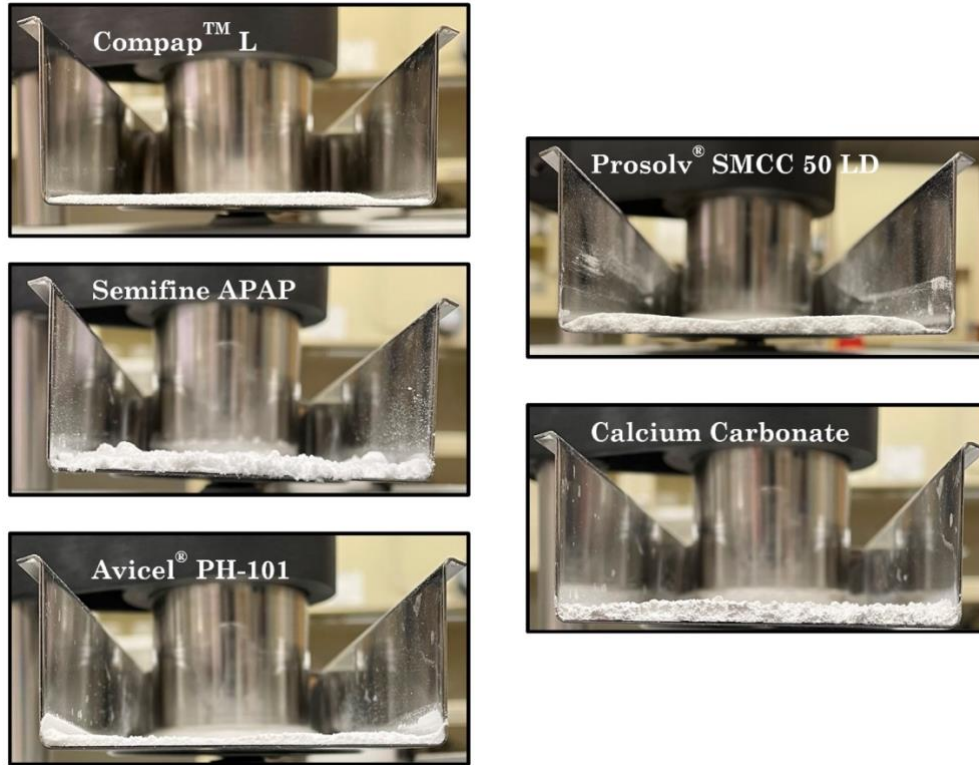


Figure 4.10: Flow layer at chute angle 10° , chute width 75 mm, chute length 210 mm, vibrational intensity 40 and at Gap of 4 mm for Compap[™] L, 12 mm for Semifine APAP, 8.5 mm for Avicel[®] PH-101, 8 mm for Prosolv[®] SMCC 50 LD, 13 mm for Calcium Carbonate.

Table 4.3: Required Gap and estimated holdup for all materials

Material	Required Gap (mm)	Estimated holdup (g)
Compap [™] L	4	6.50
Semifine APAP	12	5.23
Avicel [®] PH-101	8.5	5.25
Prosol [®] SMCC 50 LD	8	5.10
Calcium Carbonate	13	13.20

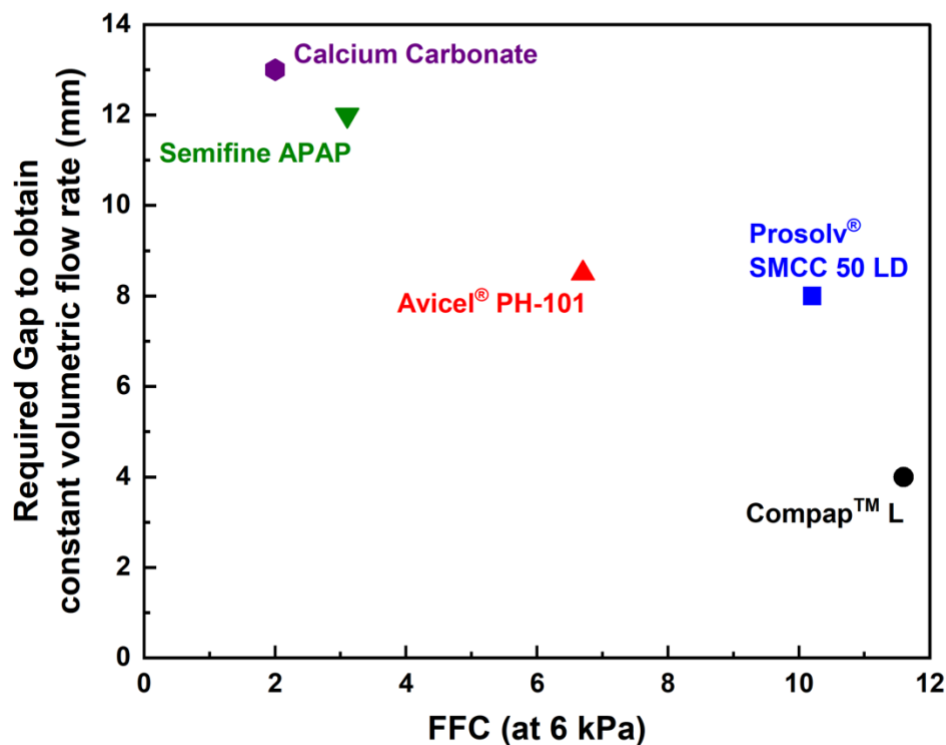


Figure 4.11: Required Gap vs. FFC for Calcium Carbonate, Semifine APAP, Avicel® PH-101, Prosolv® SMCC 50 LD, and Compap™ L at chute width 75 mm, chute angle 10°, chute length 210 mm, and vibrational intensity 40.

Compap™ L, Prosolv® SMCC 50 LD, and Calcium Carbonate. Figure 4.9 shows the graphs of holdup vs. Gap for all five materials. The Gap is kept at 0.4 for Compap™ L, the material with the highest flow function coefficient out of the five materials, and the volume of the flow layer is fixed. The estimated holdup and required Gap for all five materials are calculated at that fixed volume and shown in Table 4.3. The holdup values are verified with experiments, and Figure 4.10 shows the flow layer for all materials. Figure 4.11 shows the relation between FFC and Gap. It should be noted that the Gap can be

estimated for different materials with known FFC. It is noticed that the Gap required is higher when FFC is lower.

4.4 Conclusion

In this final chapter, a setup to measure the charge density of materials is presented. Unlike the setups described in previous chapters, the setup in this chapter uses a hopper for material input in the vibratory feeder and is proved to have better precision. The experiments performed in this chapter are carried out on different days and at different times with similar experimental conditions. Variation of angle of chute, vibration intensity level is studied. It is concluded that at a 10° chute angle, the experiments have better replicability. The distance between hopper opening and chute (Gap) is also studied in this chapter. The Gap is helpful to control the volume of the powder flow layer and make sure the flow layer has similar thickness across the length. Therefore, to compare charge density between different materials, the flow layer volume can be kept constant. That is possible by varying the Gap, for different materials in Setup C. It is noted that the length and width of the chute will be constant for all the materials. Five different materials are compared. It is concluded that FFC can be used to estimate Gap while comparison different materials at constant powder flow layer volume. The final methodology to measure the charge density of multiple materials compared to each other is presented in the next chapter.

Chapter 5: Methodology for Powder Tribocharging Characterization

The methodology to measure powder tribocharging and to compare multiple materials is proposed in this chapter. The method is based on Setup C, presented in Chapter 4 of this work.

Steps for powder conditioning for the experiment are as follows:

- (i) Powders need to be stored at desired humidity for 48 hours in a humidity chamber at ambient temperature.
- (ii) Powder containers should be stored in a manner of one container per experiment to minimize variation.

To compare different materials, the volume of the powder flow layer for all materials needs to be constant. For calculation of the constant volume, calculate the suitable Gap value for each material. The steps to calculate Gap are as follows:

- (iii) Calculate holdup at multiple Gap values using Setup C (The holdup is calculated as the weight of the material present on the chute for a particular Gap value when the flow is in the equilibrium).
- (iv) Using the tapped density of the material and holdup, find the volume of material present in the powder layer.
- (v) Repeat steps (iii) and (iv) for each material in the comparison study.

- (vi) Fix the volume of powder present in the flow layer to compare all material at that volume.
- (vii) For the fixed volume, find Gap and holdup values for all materials (using the data obtained from step (iii)).
- (viii) To verify the holdup from step (vii), experiment with the Gap from step (vii), repeat this for all materials included in the study, and list required Gaps for all materials.

Experimental procedure:

- (ix) The humidity of the experimental setup chamber should be the same as the desired humidity, at least 1 hour before the experiment time.
- (x) The hopper, stainless-steel cup, vibratory feeder chute, and faraday cage should be clean and dried, in addition to the grounding of the hopper and the vibratory feeder chute.
- (xi) Keep the vibrational feeder Retsch DR100 at 10° of inclination level with a fixed vibration amplitude (40% of the maximum recommended)
- (xii) Adjust the Gap according to the value decided from step (viii).
- (xiii) Experiment at decided Gap, vibration level, and at 10° chute angle.
- (xiv) Repeat the experiment at least three times for each material.

Conclusion

In this work, the method to measure powder tribocharging is presented, which is suitable for characterizing different granular materials under identical volumetric flow rates. Commercially available electric charge analyzer GranuCharge™ is studied for accuracy. In general, it was observed that GranuCharge™ has significant variability in experiments performed on different days with the same conditions, for Avicel® PH-101, Avicel® PH-102, Avicel® PH-105, Semifine APAP, and Powder APAP. In addition, the mass flow rate is not constant along with a test, and significant powder adhesion is observed in cohesive materials. The feeder-channel-faraday cup setup is developed to have more control over the flow conditions.

The setup with a channel where the powders slide down by gravity and get tribocharged is proposed. The effect of vibrational amplitude of feeder, angle of inclination of feeder chute, the length, and angle of inclination of the channel on mass flow rate and charging tendency is studied. It is observed that the flow of materials is not controlled on the channel, as the channel is non-vibrating. Random adhesion on the channel affects the mass flow rate and charging behavior. Moreover, cohesive materials do not flow on the channel with just gravitational force. In conclusion, setup is challenging to standardize due to the lack of sufficient control on the flow pattern of the powder. In addition, it is found that the residence time of material on the surface, the mass

flow rate, and volumetric flow rate are important factors for the characterization of powder tribocharging.

The final methodology is developed with a vibrational chute and Faraday cup. This setup can control the flow pattern of the free-flowing and cohesive type of materials. A hopper is used to feed the material to the vibrating channel. The hopper and designate the powder input and spread on the chute, which decreased the manual feeding errors. The volumetric flow rate can be controlled by varying the distance between hopper and chute (Gap), for each material, according to their flowability. Experiments showing the dependency of charge acquisition on the volume of the flow layer are presented. For Avicel® PH-101, it is observed that for lower Gap, higher charge acquisition is possible.

It is proposed that to exhibit a significant comparison of tribocharging between different materials, the volumetric flow rate should be kept constant. This methodology is suitable for comparing charge gain between materials with different flow behavior. A relation between the flowability coefficient and the hopper position was established to estimate suitable Gap values for different materials. The charge measurement experiments using the methodology presented are the focus of the current work. The method is also used to include tribocharging characterization of materials to the material property database generated in the group by considering flow properties and particle size distribution of powders. Future work includes tribocharging

characterization of powder mixtures and observing the effect of different parameters such as relative humidity and contact surface on tribocharging of single powder and powder mixtures.

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