One-step Measurements of Powder Resistivity as a Function of Moisture

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***Abstract*—A new one-step technique to measure the effect of humidity on powder resistivity has been previously presented. In this technique, humidity of surrounding air is rapidly changed either from low to high or from high to low. Resistivity is measured simultaneously with the diffusion of water molecules. System consists of an electrode arrangement which generates a uniform electric field in front of the counter electrode regardless of moisture gradient. In this presentation, we provide more experimental data obtained with five different powders, two lactoses, monoammonium phosphate, sodium chloride and common sugar. One-step measurements and traditional multi-step measurements have been performed. Also, additional measurements have been performed using standard resistivity cell. Results are compared and it can be concluded that the new technique provides meaningful results although significant hysteresis was observed during humidification and drying cycles. Finally, charging of the powder was also measured and it was noticed that it decreases with decreasing resistivity and increasing humidity.**

I. INTRODUCTION

Resistivity measurements of powders are important since the resistivity plays a significant role in powder triboelectrification. It is generally thought that decreasing resistivity increases charging. This due to the fact that the charge which is transferred in a contact, is able to flow back if the powder resistivity is low. Excess charging can cause a lot of problems in different powder handling operations, such as mixing, filling and transport. Charging experiments can be performed in many different ways, for example, sliding a sample down into a Faraday cup via a pipe, or by charging in fluidized bed device [1-3]. In general, charging experiments suffer from quite poor reproducibility and are difficult to perform consistently in a normal laboratory without specially trained personnel. Resistivity measurements, on the other hand, are quite easy to perform compared to charging experiments.

It is well known that increasing the humidity of surrounding air decreases powder resistivity due to adsorbed water layer on the particles. Thus, increasing the humidity also decreases charging [4-5]. Thus, humidification is often used to control the charging. However, there are several situations where humidification can cause additional problems. For example, in pharmaceutical industry powders are processed in many unit operations and the particles will charge. Humidification could reduce the problems but some of the materials are chemically and/or physically stable only in dry atmosphere. In chemical industry, large volumes of powders can be transferred pneumatically and high charges may build up. Again humidification would reduce the charging but excess water may significantly increase cohesion and hamper powder flow.

According to the previous discussion, there is a need for a method for determining a humidification level which is high enough for charge control but low enough not to cause additional problems.

Traditionally, the relation between resistivity and humidity has been measured in several steps. Dry powder is placed in a resistivity cell, voltage is applied between the electrodes and the current is recorded. From the recorded voltage and current, resistance can be calculated using Ohm's law. For the calculation of resistivity of the material, dimensions of the cell need to be known. The volume of standard resistivity cell is roughly 0.5 liters. Electrodes are circular, placed vertically and their radius is 25 mm. Spacing between the electrodes is 5 mm and the electrodes are aligned parallel to each other [7]. Next, the humidity of surrounding air is increased to a certain value, typical increase being approximately 10 – 15 RH%. Water molecules will then diffuse into the powder bed. Depending on the powder properties, particle size and the height of the powder bed, the diffusion will last for a certain time. For many powders, the time needed for the system to reach equilibrium can be several days. The equilibrium has been reached when the current signal has reached a steady state. Then humidity is increased again in a stepwise manner. So, measurement of the whole range of interest can take weeks.

In this work, a one-step method was used together with a new resistivity cell geometry. In this method, humidity is rapidly changed from either low to high, or high to low. The humidity is measured at the height of the electrodes simultaneously with resistivity. The procedure and electrode geometry are presented in more detail in previous paper [8] and is only briefly discussed here. Electrode maintained at high potential is square and the current electrode is narrow and aligned horizontally. The current electrode is surrounded by a square shaped and grounded guard plate which prevents current which flows through more humid areas from reaching the current electrode.

Resistivity measurements were performed on five different powders between relative humidities of approximately 10 % to 90 % (10% to 65% on more conducting samples). Results were compared with measurements performed with a standard cell and multi-step humidification. Since the objective of the humidification is to reduce the charging, charging measurements were also performed at different humidities. Charging was performed using a small fluid bed device with closed air loop.

II. MATERIALS AND METHODS

A. Materials

Five different powders were chosen for the study. Materials were chosen so that they cover a large resistivity range from 10^5 to 10^{14} Ωm. Two different grades of lactose α-monohydrate were used: Capsulac 60 and Prismalac 40 (Meggle, Germany). Common sugar was also used (Suomen Sokeri Oy, Finland). Monoammonium phosphate (Krista-MAP) was obtained from Yara, Belgium and Sodium chloride from Merck, Germany.

B. Methods

1) Resistivity cells

New resistivity cell and the measurement procedure are presented in detail in previous article [8]. Schematic image of the system is presented in Fig. 1. Cell constant *K* of the new resistivity cell was 0.0375 m.

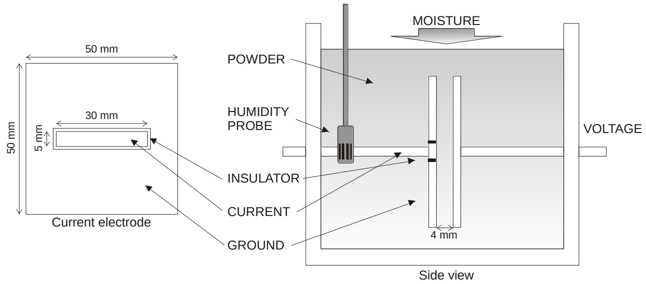

Fig. 1. Schematic image of the resistivity cell. Figure on the left shows the cross section of the current electrode and guard plate. Figure on the right represents a side view of the device.

Another resistivity cell was also used and it has been built according to standard [7]. It has circular electrodes with 50 mm diameter and spacing 5 mm which results in a cell constant *K* of 0.393 m. Resistivity cells were placed inside a sealed and grounded chamber. Predetermined relative humidity inside the chamber was obtained using saturated salt solutions and dry air flow. Humidities of the surrounding air and within the powder were measured using HMI38-instrument (Vaisala) equipped with two probes, HMP37E and HMP35E (Vaisala). Resistance across the electrodes was measured using an electrometer (Keithley 6517A, Keithley Instruments Inc.). Voltage across the electrodes was kept constant through the whole drying/humidification cycle and it was set to 1000 V for both lactoses and sugar, 100 V for MAP and 10 V for NaCl. Humidity inside the powder at the height of the current electrode, humidity of surrounding air, and measured resistance *R* were recorded simultaneously using a virtual instrument written in LabVIEW and a PC. Resistivity ρ of the sample was calculated from measured resistance *R* using equation $\rho = KR$.

One-step measurements were performed so that the resistivity cell containing the sample was placed inside the chamber and the chamber was flushed with dry air. When the relative humidity inside the powder was approximately 10 %, a petri dish containing water (or saturated salt solution in some cases) was placed inside the chamber. The voltage was turned on and the values were recorded until the relative humidity inside the powder was approximately 90 % (65 % using MAP or NaCl). Then, petri dish was removed and the chamber was flushed with dry air. Data was recorded during the drying cycle until the powder was again approximately 10 % dry.

Multi-step measurements were performed in a similar way. Saturated salt solutions were used to obtain constant relative humidity. Data was recorded until resistivity signal had reached a steady-state value. Then, another salt solution was introduced into the chamber and the procedure was repeated until the whole humidity range had been covered.

During the measurements it was noticed that the resistivities of dry lactoses and sugar were too high for accurate measurement. Thus, resistivities above 10^{14} Ωm could not be recorded and has to be considered unrealiable.

2) Charging

Samples were charged by fluidizing a predetermined amount of sample in a stainless steel column (inner diameter 36 mm) for 5 minutes. The air distributor at the bottom of the column was made of a fine stainless steel mesh. Prior to measurements, the column was washed with tap water and a brush, rinsed with purified water, rinsed with ethanol and blow-dried using clean air.

Then, column was assembled and the air circulation was turned on. Air flow velocity was monitored using a hot wire anemometer (Envic AFD-1) and the humidity using a dew point meter (Easidew EA2-TX-100-HD, Michell Instruments). Air circulation in the system followed a closed loop. Dry relative humidity conditions were adjusted by flushing the system with dry air. Humid conditions were obtained using humidified air. Since the loop was sealed, relative humidity remained constant during the charging. When a predetermined relative humidity was reached, the blower was switched off. Sample was then poured into the column under an AC neutralizer and the blower was turned on. Beginning of the fluidization was observed by a small induction probe which measured current signal a few millimeters above the air distributor. When the powder had fluidized for 5 minutes, air flow was stopped, air distributor was moved aside using a handle and the sample dropped directly into a Faraday cup where its charge was measured using an electrometer (Keithley 6514, Keithley Instruments Inc.). Sample was then removed, weighted and specific charge (nC/g) calculated. The charging measurements were repeated three times for each powder and humidity. All data was recorded and the device controlled by a virtual instrument written in LabVIEW.

Since the samples had somewhat different properties, fluidization parameters were also different. Optimal fluidization air velocity was obtained by visual observation of fluidization behavior in a transparent acrylic column. Table 1 presents fluidization air velocities and sample masses for different powders.

TABLE 1. FLUIDIZATION AIR FLOW VELOCITIES AND SAMPLE MASSES FOR DIFFERENT POWDERS

Sample	v [m/s]	m [g]
Prismalac 40	0.62	10
Capsulac 60	0.48	10
Sugar	0.62	15
NaCl	0.81	15
MAP	0.83	10

III. RESULTS

A. Resistivity as a function of relative humidity

As mentioned in the previous paper, one-step measurements suffer from a significant hysteresis loop during humidification and drying cycles. Starting from dry condition, when the humidity of surrounding air was instantaneously increased to high, resistance across the sample decreases rapidly at first and then slows down as the measurement and diffusion of moisture into the powder progresses. Similarly, when high humidity was changed to dry, resistance usually increases very rapidly at first. The size and shape of these hysteresis loops might look different for different powders. As an example of this, Figure 2 shows humidification-drying cycles of Capsulac 60 and NaCl.

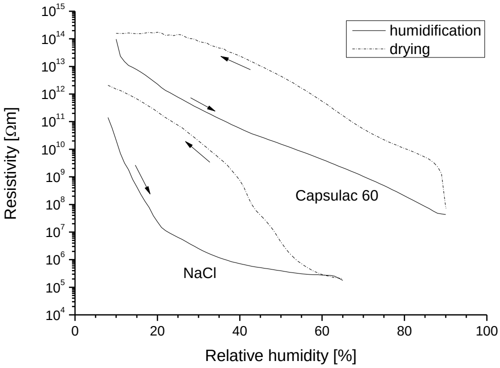

Fig. 2. Resistivity of Capsulac 60 and sodium chloride as a function of relative humidity. Data obtained using one-step method and the new resistivity cell. Samples were first humidified and then dried. Note the logarithmic scale.

Since the difference between resistivity during humidification and drying at certain humidity can differ two orders of magnitude, it is quite impossible to tell what the real (steady-state) value would be. Therefore, multi-step measurements were also performed. In these measurements, humidity (and resistance) was allowed to stabilize before the next value of humidity was set. It should be emphasized at this point that one-step measurement takes approximately two days while multi-step measurements take four weeks.

It was assumed that resistivity values obtained using multi-step measurements would be in the middle of the hysteresis loop. However, it was noticed that this is not always the case. Taken all the measurements in consideration, results can be summarized as follows:

- end-points of the hysteresis loops were close to steady-state values
- steady-state values were closer to the center of the loop at low humidities than at high (>50 %) humidities
- steady-state values draw relatively straight line as a function of moisture in a logarithmic scale

As an example, Figure 3 presents both one-step and multi-step data for Prismalac 40 and Figure 4 shows steady-state resistivities for MAP and both lactoses. Figure 3 shows that the steady-state values are close to the ones obtained during the humidification cycle when the relative humidity is more than 50 %. At lower humidities the steady-state values are close to the center of hysteresis loop. Figure 4 shows that steady-state resistivities form a straight line on a logarithmic scale, thus resistivity decreases exponentially as a function of moisture. Note that resistivities higher than 10^{14} Ωm could not recorded using the current setup.

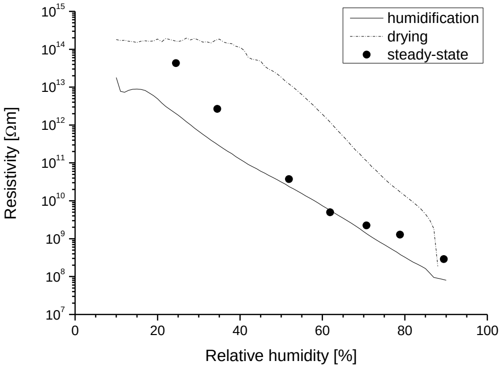

Fig. 3. Resistivity of Prismalac 40 as a function of relative humidity. Lines were obtained using one-step method and the new resistivity cell. Points present steady-state values obtained using the new cell and multi-step method.

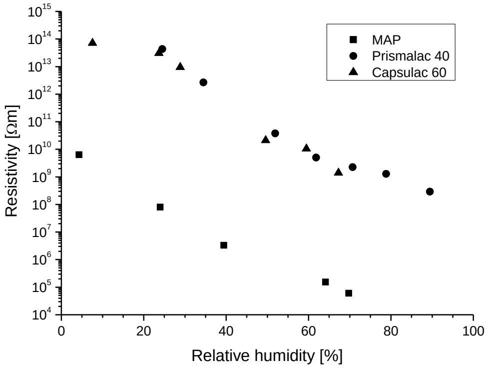

Fig. 4. Steady-state resistivities of monoammonium phosphate, Prismalac 40 and Capsulac 60 as a function relative humidity. Data obtained using new resistivity cell geometry.

For tribocharging point of view, resistivities at low humidities were considered most important. It was also noticed that steady-state resistivities located usually in the middle of hysteresis loops especially at low humidities. Thus, it was decided that the results of one-step measurements can be presented as geometric mean values, and these geometric mean resistivities are presented in Fig. 5. Figure shows that resistivity decreased as a function of relative humidity for all samples. Sodium chloride and monoammonium phosphate were most conductive. For these samples, humidification was stopped at 65 % due to high conductivity and dissolution issues.

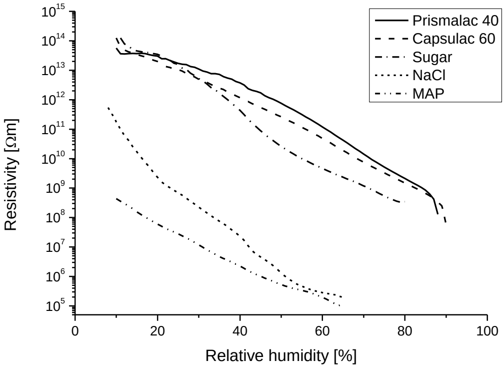

Fig. 5. Resistivities of samples as a function of relative humidity. Presented curves are calculated geometric mean values of one-step measurements.

Steady-state values which were obtained using the new cell geometry were compared with steady-state values measured using standard cell. Results for Prismalac 40 are presented in Figure 6 and it can be concluded that resistivity values are similar especially at lower humidities which are considered most important from the charging point of view.

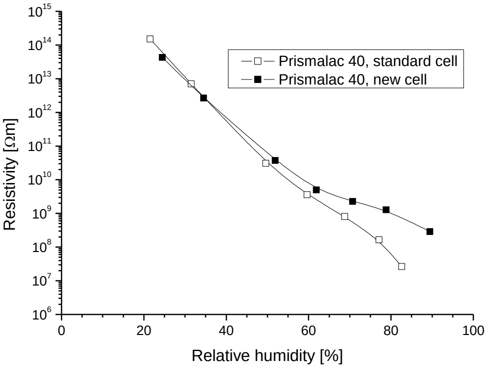

Fig. 6. Steady-state resistivities of Prismalac 40 measured using both new and standard cell as a function relative humidity.

Resistivities presented in Fig. 6 show that measured values differ at high humidities where the resistivity is low. The observed lower resistivity using standard cell could be explained by stray currents. The electrode geometry of the new cell prevents stray currents from reaching the current electrode. It is possible that when the standard cell was used, significant amount of current was able to flow to the current electrode via path outside the volume between the electrodes.

B. Charging as a function of relative humidity

Powder samples were charged according to procedure presented in chapter II.B.2. Results are presented in Figure 7.

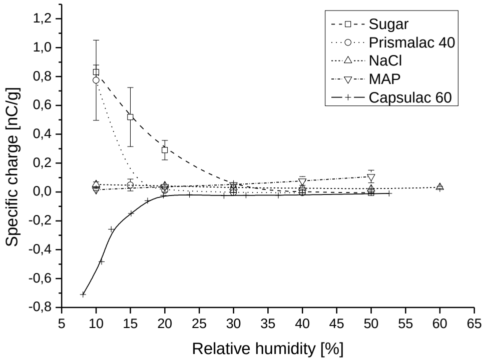

Fig. 7. Specific charge of studied powders as a function of relative humidity.

Figure 7. shows that all powders acquired a positive charge in a stainless steel column except Capsulac 60 which charged negatively. Sugar charged most (0.83 ± 0.05 nC/g) and sodium chloride and monoammonium phosphate did not acquire notable charge. It was noticed that charging decreased as a function relative humidity. There was one exception, however: monoammonium phosphate charged least at 10 RH% (0.016 ± 0.006 nC/g) and as humidity increased to 50 RH%, charging also increased to 0.107 ± 0.044 nC/g. Absolute values of specific charges of sugar and both lactoses were quite similar, as were the resistivities. Also, MAP and NaCl were most conducting and their charges were negligible. Clearly there was a trend between resistivity and generated specific charge. To study this, specific charges were also plotted as a function of resistivity and this data is presented in Figure 8.

Figure 8 shows that charging was small for all samples when their resistivity was less than 10^{13} Ωm. Charge decreased further as the resistivity decreased as can be seen from the curve of NaCl, for example. Again, monoammonium phosphate was an exception and its charging was highest at low resistivity. Reason for this is unclear, but it might have something to do with electrochemical effects at higher relative humidities.

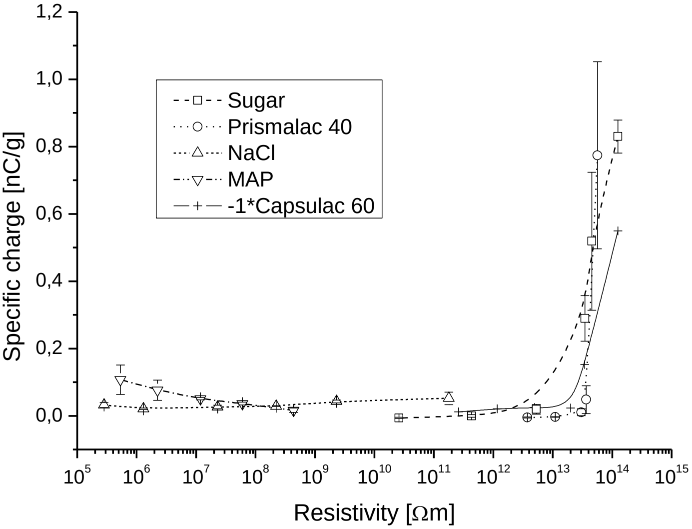

Fig. 8. Specific charges of powder samples as a function of resistivity. Absolute value of Capsulac 60 is presented in this figure.

C. Discussion

New electrode geometry was designed to enable much faster resistivity scans over a wide humidity range. The system suffers from significant hysteresis loop and determining the correct resistivity value proved to be challenging. In this work, geometric mean value of humidification and drying cycles was chosen. It seems to give quite comparable values compared to “correct” resistivities measured using steady-state methods, especially at relative humidities below 40 %. Minimum and maximum values of resistivity measured using one-step method correlate relatively well with steady-state values, which can be considered natural, since ends of drying and humidification cycles are essentially same as steady-state measurements. Resistivity was found to decrease exponentially with moisture, i.e. plots were quite linear in logarithmic scale. Thus, it could be tempting to extrapolate whole humidity range by measuring the end-points only. However, some materials might have discontinuities, such as phase transitions, at certain humidities. For this reason, the measurement method should be further developed.

Results show that increasing humidity decreases resistivity which in turn seems to decrease measured specific charge. One might ask whether excess moisture actually reduces charging or does it simply increase charge decay. In this study, particles charged in contact with stainless steel column and air distributor mesh. Then, after five minutes air flow was switched off and the particles became at rest on the bottom of the column. When the particles are resting charge is able to flow back to grounded steel column. Thus, time period between switching off the fan and dropping the powder into the Faraday cup could have a significant effect on the charging results. In this study it was necessary to wait until the air flow was below 0.2 m/s, otherwise the powder would not fall. Since different fluidization velocities were used, this time period also differed slightly from sample to sample. If it assumed that charge decays exponentially according to Equation 1 [9], relative permittivity of the powder is 2 and it takes 10 seconds to drop the powder to Faraday cup, it follows that 10% of charge has decayed when resistivity is $5\cdot10^{12}\Omega$m.

$$q = q_0 e^{-\frac{t}{\rho\varepsilon}} \tag{1}$$

In this study, the time period in which the particles were at rest, was approximately 5 seconds. Thus, the effect of this charge decay becomes important when the resistivity is less than approximately 10^{12} Ωm.

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