

# The electrical resistivity and relative permittivity of binary powder mixtures

Janne Peltonen<sup>a,\*</sup>, Matti Murtomaa<sup>a</sup>, Kelly Robinson<sup>b</sup>, Jarno Salonen<sup>a</sup>

<sup>a</sup>*Department of Physics and Astronomy, University of Turku, 20014 Turku, Finland*

<sup>b</sup>*Electrostatic Answers, LLC., 15 Piping Rock Run, Rochester, NY 14450, USA*

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## Abstract

Effective electrical resistivities and static relative permittivities of binary powder mixtures were measured and compared to theoretical equations given by Generalized Mixture Rule (GMR). The measured values for mixtures were influenced by the resistivity and permittivity of the pure component materials, the particle sizes and by the sticking between the different particles. According to the Random Model (RM), the effective resistivity and relative permittivity of any randomly packed mixture material can be calculated from the component resistivities and relative permittivities, respectively, and their concentrations. It was shown that RM gave good predictions for both quantities for powders if the particle sizes of the component materials were almost the same. However, this was not the case when large NaCl particles were mixed with fine sugar particles. When these mixtures were prepared, finer, more numerous sugar particles coated the larger NaCl particles, and the mixtures were not random but closer to the series configuration. Effect of particle size was studied briefly by mixing very fine NaCl particles with

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\*Corresponding author. Tel.: +358 2 333 5694

*Email address:* `janne.m.peltonen@utu.fi` (Janne Peltonen)

sugar of similar size. With these mixtures, results predicted by RM were obtained.

*Keywords:* electrical resistivity, relative permittivity, powder mixtures, dielectric properties, electrostatics, random model

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## 1. Introduction

Electrical resistivity and relative permittivity are fundamental physical quantities of insulating materials. Volume resistivity is a property of a material that quantifies its ability to resist a flow of electric current. It is a crucial parameter for a powder material in powder handling processes, for instance in pharmaceutical or food industry, since it is related to electrostatic charging (triboelectrification) of powders and to dissipation of accumulated charge. Therefore, it can be used, at least to some extent, to estimate the time required for the charge to decay to ground [1, 2, 3]. Relative permittivity, or dielectric constant, of a material expresses how much an electric field decreases inside the material compared to vacuum. Thus, it is a measure of the polarizability of the material. The relative permittivity is a function of frequency of the applied electric field. It is known as static relative permittivity, or static dielectric constant, if the frequency of the field is zero. Relative permittivity is to some extent related to resistivity [1].

Resistivities of powder mixtures have been studied surprisingly rarely, taking into account that powder mixtures often charge in a more complex and unpredictable manner than pure materials [4, 5, 6, 7]. Murtomaa and Laine [4] showed that the resistivity of lactose–glucose mixture followed a linear relationship with mass concentration. However, measurements were

only performed using these two powders, and the resistivities of the pure materials were of the same order of magnitude. Thus, the linear behavior cannot be generalized for all powder mixtures.

The resistivity of a powder is commonly measured using a parallel-plate cell. The powder is poured between two parallel electrodes, potential difference is generated across the electrodes, and the electric current is measured [1]. Parallel plate electrodes can also be used to measure the relative permittivity. The electrodes and the medium between them form a capacitor, which capacitance can be measured. The capacitance is directly proportional to the relative permittivity.

In this study, resistivity and static relative permittivity of powder mixtures were studied with powders with resistivities of different order of magnitude. Measurements were performed at low humidities since humidity has a major influence on the resistivity [8, 9, 10]. The results were compared to equations given by the Generalized Mixture Rule [11, 12, 13]. To increase reproducibility in resistivity measurements, the powders were packed between the electrodes using a constant force produced by a spring.

The obtained results can be used, for instance, to study how randomly powder mixtures are packed. Second, it can also be useful if some powder's resistivity is too high to be measured reliably in a traditional way: the powder could be mixed to some other material with much lower resistivity. Measurements could be made for mixtures with different concentrations. Then, the resistivity of the powder in pure form could be obtained by fitting the best fitting curve described in section 2 and then by extrapolating into percentage value of 100 %. In addition, if the resistivities and particle sizes of the pure

component materials are known, the volume fraction of a mixture could be calculated by measuring the resistivity of the mixture.

## 2. Some theoretical approaches

### 2.1. Resistivity

#### 2.1.1. Ordered mixtures

The electrical resistivity  $\rho$  of a material is defined as

$$\rho = R \frac{A}{l}, \quad (1)$$

where  $R$  is the resistance,  $A$  is the cross-sectional area and  $l$  is the length of a sample. If the resistivity is measured using a parallel plate resistivity cell, the  $A$  is the surface area of the electrodes and  $l$  is the spacing between them. For material mixtures, powders or bulks, the measured resistivity (effective resistivity) depends significantly on the arrangement of the component materials between the electrodes. Two most simple binary mixture structures are presented in Fig. 1 (shown in 2D for comfort). In Fig. 1a., the components are layered so that they are in connected in series. If  $\rho_1$  and  $\rho_2$  are the resistivities and  $\chi_1$  and  $\chi_2$  the volume fractions (so that  $\chi_1 + \chi_2 = 1$ ) of the phases 1 and 2, respectively, the effective resistivity  $\rho_{\text{eff}}$  can be obtained from equation

$$\rho_{\text{eff}} = \chi_1 \rho_1 + \chi_2 \rho_2. \quad (2)$$

In this configuration, the effective resistivity is dominated by the more resistive phase.

In Fig. 1b., the phases are connected in parallel. The effective resistivity can be calculated using equation

$$\frac{1}{\rho_{\text{eff}}} = \frac{\chi_1}{\rho_1} + \frac{\chi_2}{\rho_2}. \quad (3)$$

In this configuration, the effective resistivity is dominated by the less resistive phase.

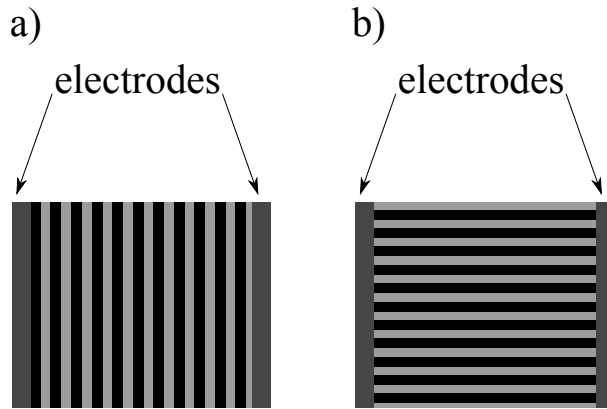


Figure 1: Binary material layered a) in series, b) in parallel. Black and grey areas between electrodes indicate different material components.

A homogeneous mixture can be obtained if the phases form a chess-board-like structure. At first, it might seem that a chess board structure is equivalent to the series geometry and that the resistivity could be calculated from equation (2). However, the case is different. The volumes of same phase are connected to each other by their edges. Therefore the current has a continuous path from one electrode to the other in the less resistive phase, and for that reason, the effective resistivity is smaller than proposed by equation (2).

The resistivity of the series geometry is the upper limit on resistivity and the resistivity of the parallel geometry is the lower limit on resistivity.

Resistivities of other configurations are between these limits.

### 2.1.2. Random mixtures

In reality the mixtures are more likely to consist of randomly distributed phases. Different models for randomly distributed volumes have been proposed (see [14] or [15], for instance). According to the Random Model (RM), a random mixture consists of randomly distributed series and parallel connections. The effective resistivity is

$$\rho_{\text{eff}} = \rho_1^{\chi_1} \rho_2^{\chi_2}. \quad (4)$$

In this configuration, neither of the phases dominates but they have an equal impact on the effective resistivity.

McLachlan et al. [14] introduced a phenomenological General Effective Media (GEM) equation to describe resistivity of binary composite materials. Ji et al. [11, 12, 13] presented a more general Generalized Mixture Rule (GMR) to predict some mechanical properties of isotropic mixture materials using the properties, volume fractions, and micro-structures of the component materials. If we use the GMR for effective resistivity, we have

$$\frac{1}{\rho_{\text{eff}}^J} = \frac{\chi_1}{\rho_1^J} + \frac{\chi_2}{\rho_2^J}. \quad (5)$$

The exponent  $J$  can be considered as a scaling coefficient that depends on the shape and distribution of the mixture components [11, 12]. The cases  $J = -1$  and  $J = 1$  correspond to the series and parallel configurations, respectively. The case  $J \rightarrow 0$  yields to RM case [11, 12]. Hence, the ordered mixtures and the RM are special cases of GMR.

## 2.2. Relative permittivity

### 2.2.1. Ordered mixtures

The capacitance  $C$  between two parallel plates with area  $A$  and separation  $d$  is given by

$$C = \epsilon_0 \epsilon_r \frac{A}{d}, \quad (6)$$

where  $\epsilon_0$  is the vacuum permittivity, and  $\epsilon_r$  is the relative permittivity of the dielectric between the electrodes. According to equation (6), capacitance is directly proportional to relative permittivity. Therefore, if  $\epsilon_1$  and  $\epsilon_2$  are relative permittivities and  $\chi_1$  and  $\chi_2$  the volume fractions (so that  $\chi_1 + \chi_2 = 1$ ) of phases 1 and 2, respectively, the effective relative permittivity  $\epsilon_{\text{eff}}$  for a binary mixture is in series geometry

$$\frac{1}{\epsilon_{\text{eff}}} = \frac{\chi_1}{\epsilon_1} + \frac{\chi_2}{\epsilon_2}, \quad (7)$$

and in parallel configuration

$$\epsilon_{\text{eff}} = \chi_1 \epsilon_1 + \chi_2 \epsilon_2. \quad (8)$$

For the effective relative permittivity, the parallel and series configurations are the upper and lower limits, respectively.

### 2.2.2. Random mixtures

The RM equation for the relative permittivity of a binary mixture is

$$\epsilon_{\text{eff}} = \epsilon_1^{\chi_1} \epsilon_2^{\chi_2}, \quad (9)$$

and GMR equation

$$\epsilon_{\text{eff}}^J = \chi_1 \epsilon_1^J + \chi_2 \epsilon_2^J. \quad (10)$$

Cases  $J = -1$  and  $J = 1$  are equivalent to the series and parallel configurations, respectively, and  $J \rightarrow 0$  yields to RM.

### 3. Materials and experimental methods

#### 3.1. Materials

Four materials with a large range of resistivity were studied: icing sugar (sucrose, Suomen sokeri Oy, Finland), potato starch (Finnamyl Oy, Finland), corn starch (Rol-Ryż Sp. z o.o., Poland) and NaCl (Merck KGaA, Germany). All powders except NaCl were used as received. To study the effect of different particle size, three samples of NaCl was produced. "Coarse" sample was produced by grounding by hand, and "fine" and "very fine" sample by using a ball mill pulverizer. Particle sizes were measured as aerosols using laser diffraction (Helos H2370, Sympatec GmbH, Germany). Particle sizes are presented in Table 1.

Sugar found to be the most resistive. Therefore, it was chosen to be mixed with other powders. Binary mixtures containing 25 wt-%, 50 wt-% and 75 wt-% of sugar were produced by stirring the ingredients together for about 5 minutes. In addition, pure samples were used: the 0 wt-% mixture refers to pure powders other than sugar, and 100 wt-% to pure sugar. Prior to the measurements, all the samples were first dried in vacuum oven in 60 °C for 24 hours and then let to cool to room temperature in vacuum. In resistivity measurements, the whole sample was introduced into the resistivity cell.

#### 3.2. True and tap densities

In order to determine the volume fractions of the samples, true densities of the materials were measured with AccuPyc 1330 pycnometer (Micromeritics Instrument Corp., USA). Measurements were repeated five times.



Table 1: Mean particle sizes and true densities of the used materials.

Sample	Particle size	True density
	d50 (d10–d90) ( $\mu\text{m}$ )	( $\text{kgm}^{-3}$ )
Sugar	27.2 (3.8–89.2)	$1592.6 \pm 0.4$
Potato starch	35.3 (17.3–58.7)	$1502.0 \pm 0.2$
Corn starch	18.8 (8.8–52.6)	$1519.0 \pm 0.3$
NaCl (coarse)	250.4 (55.6–477.7)	$2157.1 \pm 0.3$
NaCl (fine)	69.6 (14.1–180.7)	”
NaCl (very fine)	21.9 (6.4–59.4)	”

Tap densities were measured for the mixtures to see if the concentration had an affect on the packing of the mixtures. Approximately 6 ml of powder was inserted into a tube with diameter of 1.05 mm. The tube was then tapped against a bench until the height of the powder bed remained unchanged. The volume was defined indirectly from the height of the bed and the mass was obtained when the sample was weighed.

### 3.3. Resistivity measurements

The resistivity cell [4] consisted of vertically placed two electrodes made of stainless steel (height 10 mm, width 30 mm). The other electrode was fixed in place while the other one was movable. Since the measured resistivity is affected by the packing of the powder, the movable electrode was equipped with a spring so that the sample between the electrodes was compressed with a 10 N force. This value was chosen since it generated a powder density that is realistic in powder industry. In addition, the force was small enough

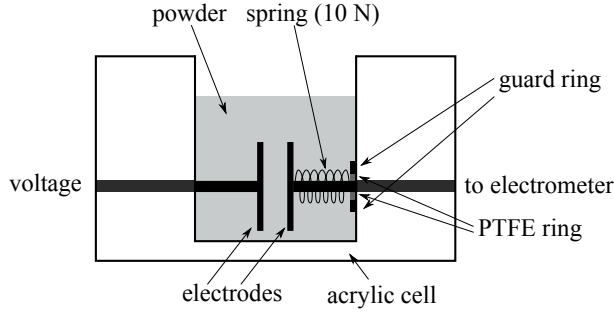


Figure 2: Side-view of the resistivity cell (not to scale).

not to modify the samples. Moreover, the same configuration has been used previously [4]. The cell itself was acrylic and its dimension were 4.0 mm x 4.0 mm x 3.6 mm (length x width x height). A grounded copper guard ring was applied around the movable electrode to prevent any possible leakage current flowing at the surface of the sample to be measured [1, 16]. The guard ring, as well as the spring, were separated from the movable electrode with PTFE. The distance between the two electrodes was measured with a built-in scale with an accuracy of 0.025 mm. A schematic image of the resistivity cell is presented in Fig. 2.

The resistivity cell was surrounded by a grounded metal chamber that acted as a Faraday cage. A dry atmosphere was produced by continually flushing the box with dry air (relative humidity less than 1 RH%). All the measurements were performed at temperature  $21 \pm 1$  °C.

A potential difference of 1000 V was generated between the electrodes with the built-in voltage source of Keithley 6517B electrometer (Keithley Instruments, USA). The voltage value was chosen so that the resulting current was high enough to be reliably measured with all the samples. The movable

electrode was grounded, and the electrometer was set to measure the resistance  $R$  between the electrodes. The resistivity  $\rho$  was then calculated from equation (1). The spacing  $l$  between the electrodes varied in the range of 0.2–2.0 mm between the measurements. When the voltage was applied, the reading on the electrometer was at first time dependent. This was caused by two factors: 1) Dielectric materials polarized in the external electric field, and generated an electric field opposite to the applied field [8]. 2) Some water molecules diffused into the powder from the surrounding air, and decreased the resistivity of the sample [9, 10]. From these reasons, the reading was taken when the signal had reached a steady state, typically after 1 or 2 days.

#### *3.4. Static relative permittivities*

Static relative permittivities were determined for each powder sample. Powder sample was inserted between two circular parallel plate electrodes (radii 20.0 mm), and the capacitance  $C_x$  of the sample was measured with Fluke 289 multimeter (Fluke Corporation, USA). The meter charged the formed capacitor with a known current for a known period of time, measured the voltage, and then calculated the capacitance. The capacitance of air filled capacitor  $C_0$  was determined with the same measurement set-up but without the powder sample while the distance between electrodes was kept the same (1 mm). The relative permittivity  $\epsilon_r$  was then calculated from equation  $\epsilon_r = C_x/C_0$ . The cell used in the resistivity measurements could have been used, but electrodes with larger surface area was used instead in order to increase measured capacitance values.

## 4. Experimental results and discussion

### 4.1. True and tap densities

The averages and standard deviations of the measured true densities are presented in Table 1. Volume proportions were calculated from predetermined mass proportions using the measured density values. Measured tap densities are shown in Fig. 3. Data points follow a linear curve with volume fraction for binary powder mixtures.

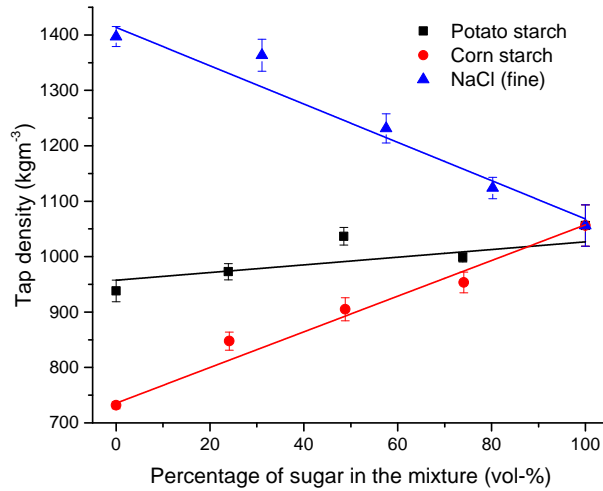


Figure 3: Tap densities of sugar mixed with NaCl, corn starch and potato starch. The curves are linear fits to the data points.

### 4.2. Resistivity measurements

The measured effective resistivities are presented in Fig. 4. The figures include GMR curves that were fitted to the data. In addition, example curves with different  $J$  values were calculated using the resistivities of the pure materials. From the fitted curves, the  $J$  values were determined (Table 2). The

measured resistivities for pure materials are presented in Table 2. The resistivities for pure NaCl samples with different particle size were somewhat different, most likely due to different morphology caused by the milling process as the samples were produced.

When sugar was mixed with corn starch, potato starch and very fine NaCl, the  $J$  value was close to zero, and therefore the results were in a good agreement with values predicted by RM (Fig. 4a–c). In these mixtures, the particle sizes of the component materials were similar. However, when larger NaCl particles (coarse and fine NaCl) were present in the mixture, the effective resistivities were larger than predicted by RM, and were closer to the series limit ( $J$  value close to  $-1$ ) than to the random resistivity (Fig. 4d–e).

Since the behavior for the mixtures that contained large NaCl particles was different compared to other mixtures, they were studied in more detail. With an optical microscope, it was observed that the finer, more numerous sugar particles coated the larger NaCl particles (Fig. 5). We speculate that the different particles became oppositely charged by triboelectrification when the mixtures were prepared causing the particles to stick. As a result, the mixtures were not randomly packed. Highly resistive small sugar particles isolated the more conducting NaCl particles from each other, and therefore there were no continuous paths of NaCl in the mixture. Thus, the effective resistivity was dominated by the sugar phase, and the structure of the mixtures were such that the effective resistivities were closer to the series configuration. When the particle sizes were similar, sugar did not coat the NaCl particles entirely, so that both NaCl and sugar formed continuous paths in the mixture. As a result, neither of the phases dominated and

results predicted by RM were obtained.

#### *4.3. Static relative permittivities*

Measured static relative permittivities are presented in Fig. 6 together with GMR curves that were fitted using the previously determined  $J$  values. The relative permittivities for pure materials are listed in Table 2. When sugar was mixed with corn starch, potato starch and very fine NaCl, the relative permittivity decreased according to RM ( $J$  close to 0) as the proportion of sugar was increased in the mixture. However, for fine NaCl–sugar mixture, the permittivity values decreased more rapidly. Just like the effective resistivity, also the relative permittivity of NaCl–sugar mixtures with large NaCl particles showed behavior related to the series configuration. Again, the explanation is that the large sugar particles were coated with smaller sugar particles so that the more continuous phase in the mixture dominated the effective relative permittivity. Since the relative permittivities of the pure materials were of the same order of magnitude, the differences between different models was small.

The samples with coarse NaCl could not be measured since the largest NaCl particles were too large to fit in between the electrodes. However, based on resistivity measurements, it is believed that the results would have been the same as with fine NaCl.

## **5. Conclusions**

Effective resistivity and static relative permittivity of powder mixtures were measured and compared to theoretical models. Random Model gave accurate predictions on both quantities if particle sizes of the component

Table 2: Measured resistivities and static relative permittivities for pure materials, and  $J$  values as they were mixed with sugar.

Sample	$\rho_{\text{eff}}$ ( $\Omega\text{m}$ )	$\epsilon_{\text{eff}}$	$J$
Sugar	$(2.3 \pm 0.5) \cdot 10^{15}$	$1.6 \pm 0.3$	–
Potato starch	$(8.2 \pm 0.8) \cdot 10^9$	$5.5 \pm 0.7$	$-0.025 \pm 0.015$
Corn starch	$(1.8 \pm 0.1) \cdot 10^{11}$	$3.6 \pm 0.5$	$-0.012 \pm 0.053$
NaCl (coarse)	$(1.1 \pm 0.1) \cdot 10^{13}$	–	$-0.775 \pm 0.060$
NaCl (fine)	$(3.2 \pm 0.2) \cdot 10^{12}$	$5.8 \pm 0.6$	$-0.815 \pm 0.037$
NaCl (very fine)	$(2.1 \pm 0.3) \cdot 10^{13}$	$5.8 \pm 0.5$	$-3.4 \cdot 10^{-4} \pm 0.300$

materials were similar. When the particle sizes were different, the larger particles were covered by the small particles of the other powder. As a result, the phase with smaller particle size was dominant. The measurements show that the structures were different for different mixtures.

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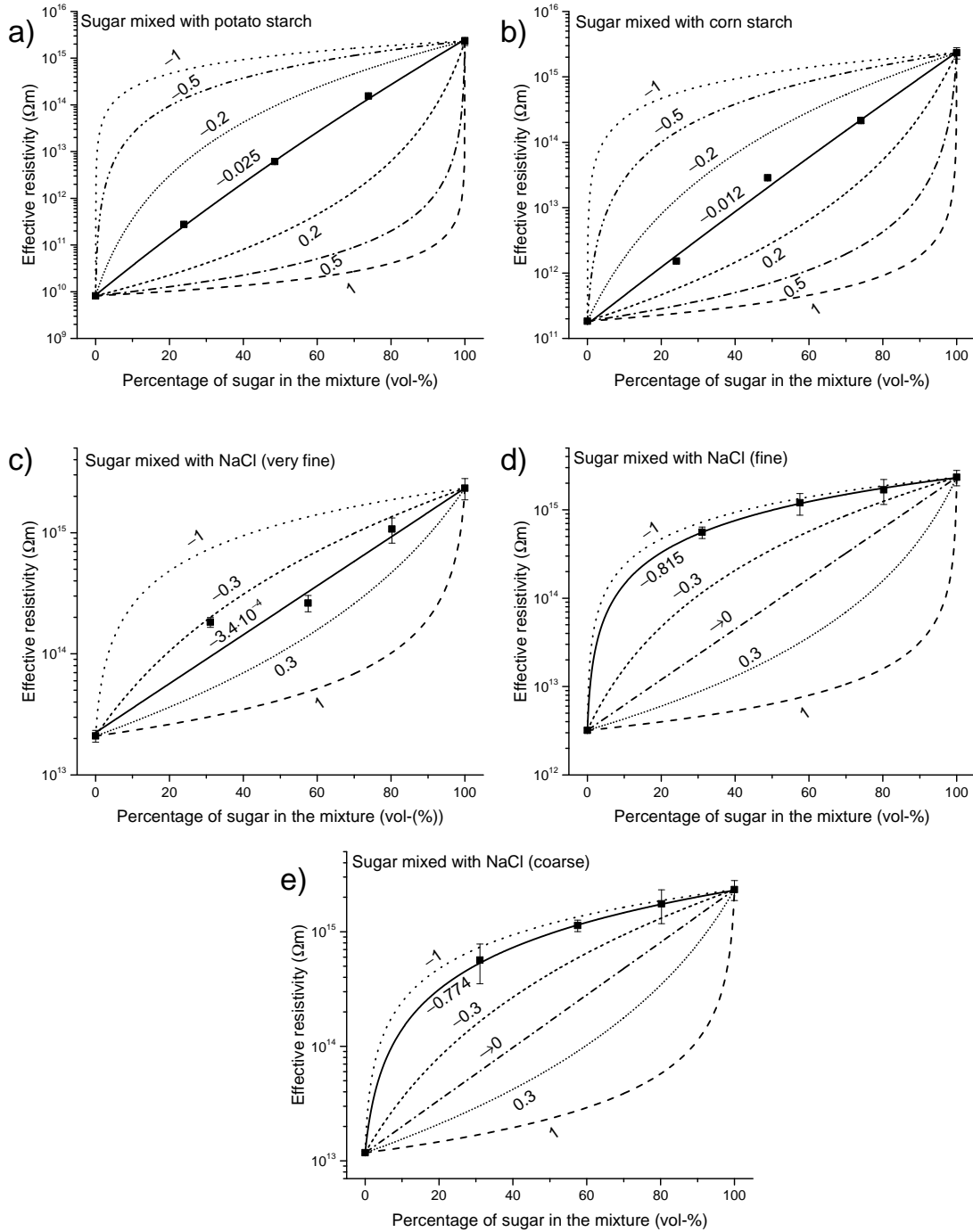


Figure 4: Measured and calculated effective resistivities as a function of proportion of sugar in a) potato starch, b) corn starch, c)–e) NaCl with different particle size. Solid curve represents the GMR curve fitted to the measured data, while dashed curves are calculated from the resistivities of the pure materials. The curves are labelled with the  $J$  value.

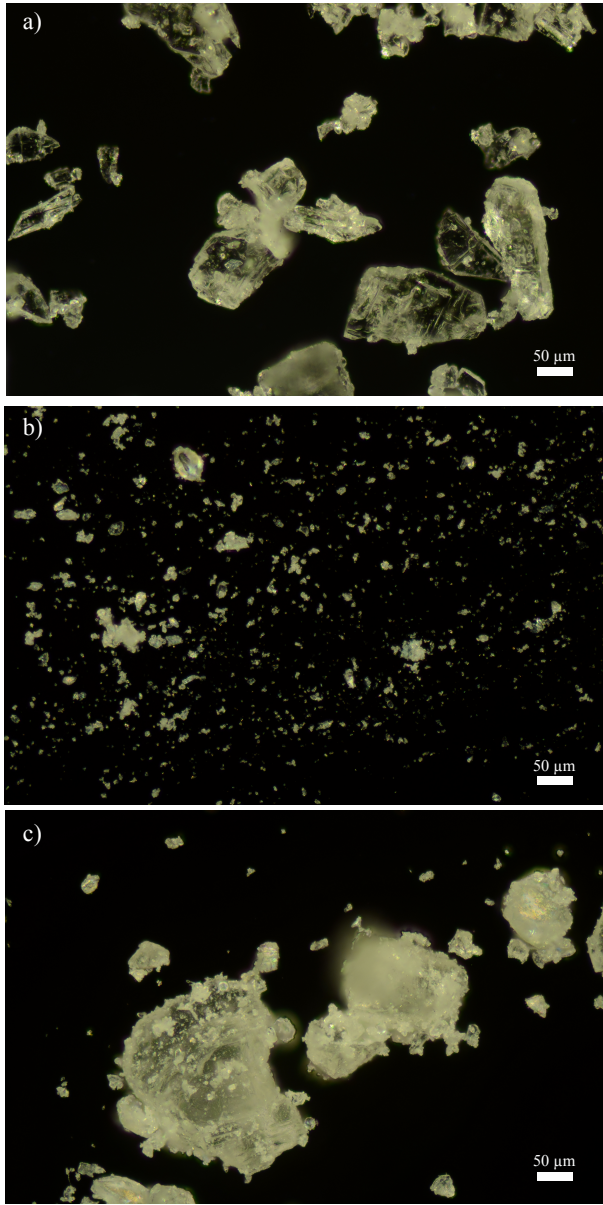


Figure 5: Microscopic images from a) NaCl, b) sugar, c) sugar coated NaCl particles.

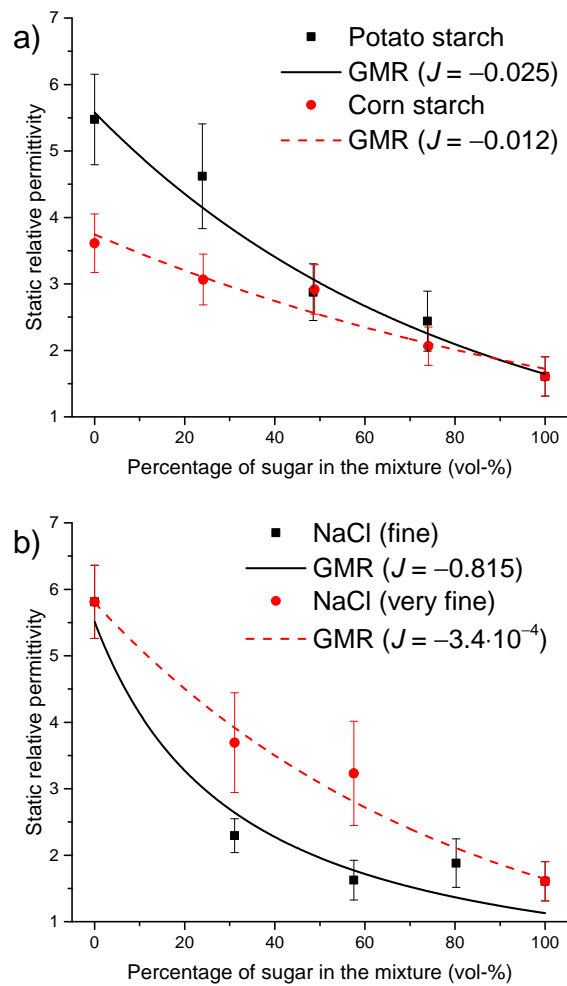


Figure 6: Measured static relative permittivities for sugar mixed with a) potato starch and corn starch, b) NaCl with different particle size. The presented curves were fitted to the data points with the determined  $J$  values.