Electrostatics and cohesion: Cause or effect?

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THE STATE UNIVERSITY OF NEW JERSEY
RUTGERS

Powder Flow 2009
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NSF

Charging and Granular Flow

- Adhesion
  - *Grains stick to surfaces*
    - *Coating*
  - *Grains stick to one another*
    - *Agglomeration*
  - *Jamming*
  - *Nonuniform/intermittent flow*
  - *Unpredictable behavior*
  - *Poor mixing*
- Repulsion
  - *From charged surfaces*
  - *From other grains*
  - *May lead to segregation*

Sand adhered to a hopper

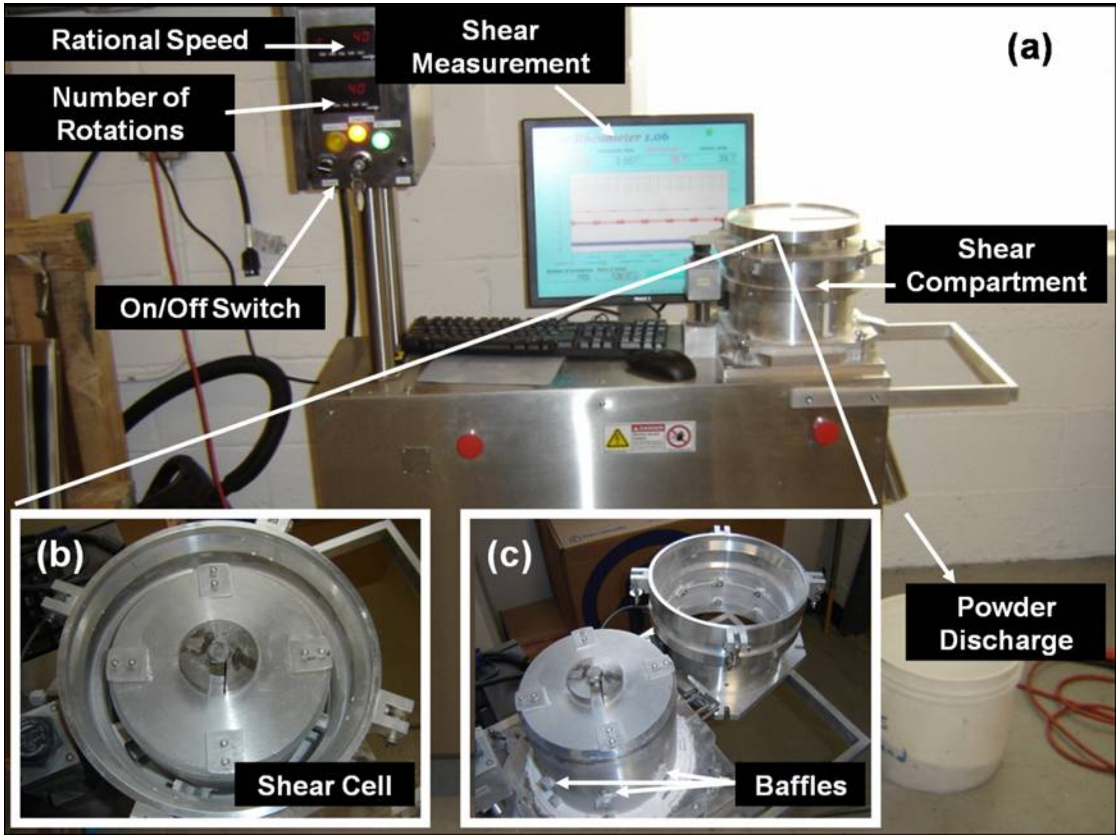
Cellulose adhered to a charged rod

Lemarche et al., Electrostatic instabilities, charging and agglomeration in flowing granular materials, 2008.

Case Study 1: Pharmaceuticals

- (1) Preblend
  - *50% Avicel 102 + 50% Pharmatose*
- (2) Preblend + API
  - *9% Mic.Acetaminophen + 45.5 % Avicel 102 + 45.5 % Pharmatose*
- (3) Preblend + API + MgSt
  - *9% Mic.Acetaminophen + 45% Avicel 102 + 45% Pharmatose + 1% MgSt*
- (4) Preblend + API + MgSt + Talc
  - *9% Mic.Acetaminophen + 44.5% Avicel 102 + 44.5% Pharmatose + 1% MgSt + 1% talc*
- (5) Preblend + API + MgSt + Cab-O-Sil
  - *9% Mic.Acetaminophen + 44.5% Avicel 102 + 44.5% Pharmatose + 1% MgSt + 1% Cab-O-Sil*
- (6) Preblend + API + MgSt + Cab-O-Sil + Talc
  - *9% Mic.Acetaminophen + 44% Avicel 102 + 44% Pharmatose + 1% MgSt + 1% Cab-O-Sil + 1% Talc*

Equipment For Sample Preparation: Controlled Shear Environment



(a) Rutgers controlled shear environment. (b) Shear compartment where the powders processed were limited to 200g per run due to the limited size. (c) The baffles provide the shear stress to the powder particles.

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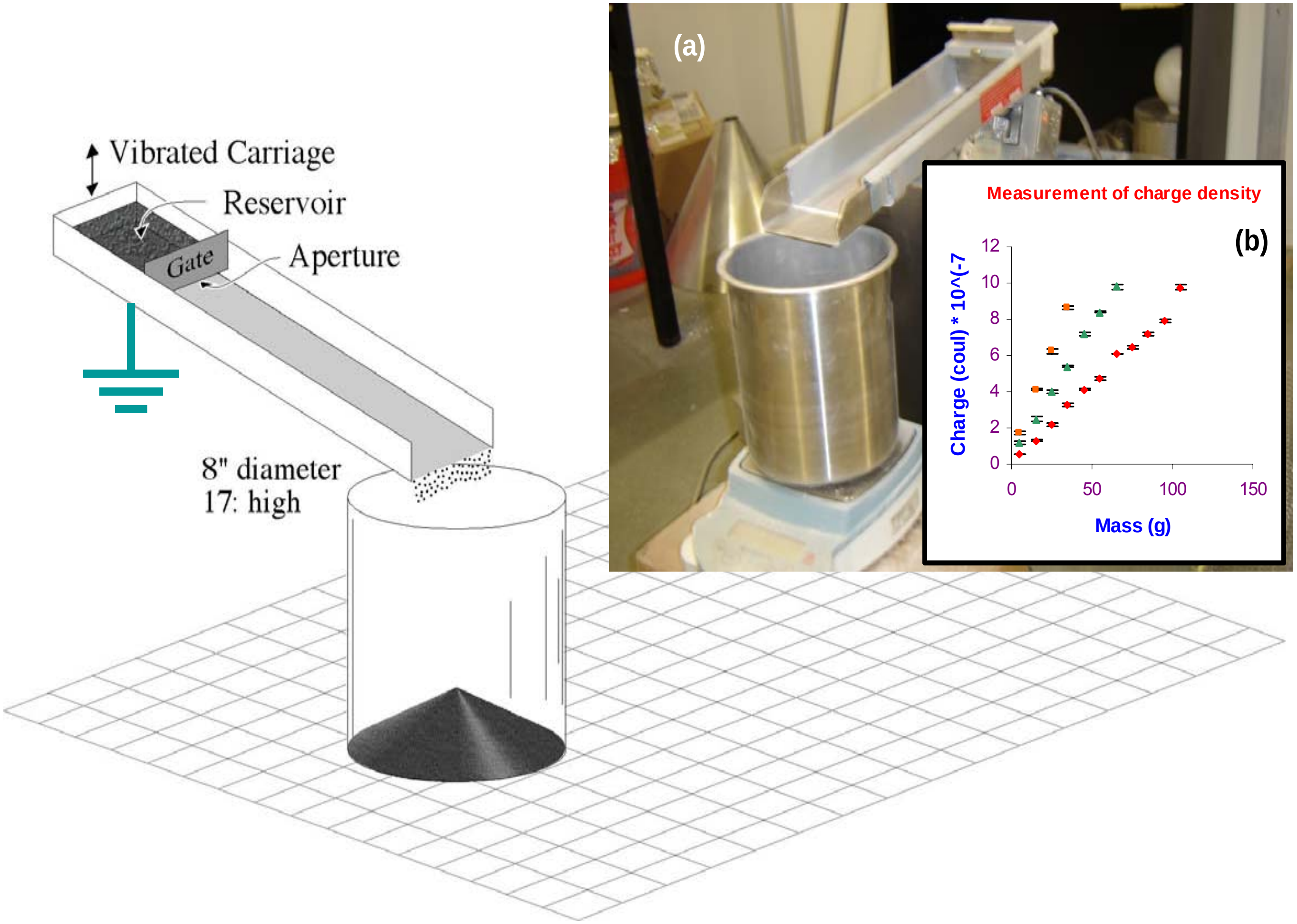
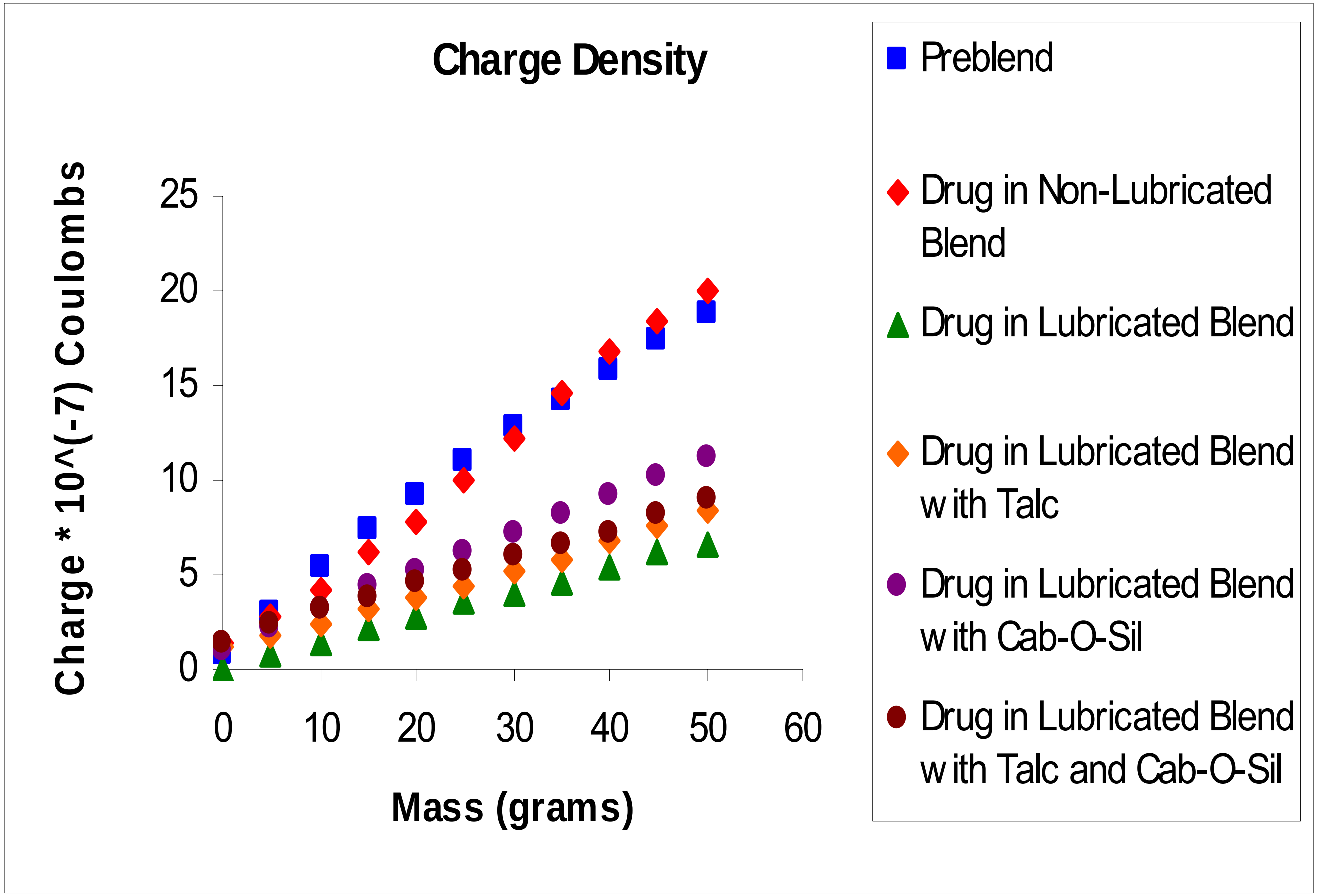
Measurement of Acquired Charge



Acquired Charge



Shear rate = 40 rpm

Strain = 640 rev

MgSt, Talc and Cab-O-Sil are effective in decreasing charge density.

Effect of Shear Rate, Strain, and Blend Composition on Electric Properties

Monopolar Charge Density

$$Y_{ijk} = \mu + B_i + R_j + BR_{ij} + S_k + BS_{ik} + RS_{jk} + \varepsilon_{ijk}$$

B_i=Blend effect

R_j=Shear Rate effect

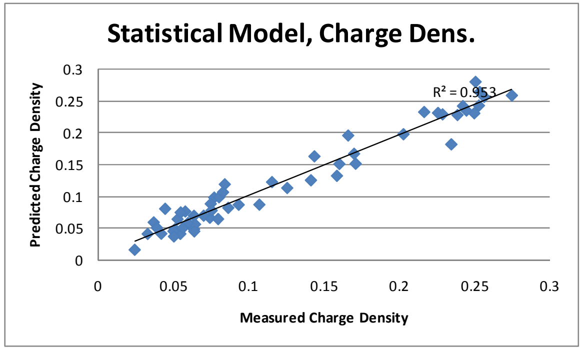
BR_{ij}=Blend-Shear Rate interaction

S_k=Strain Effect

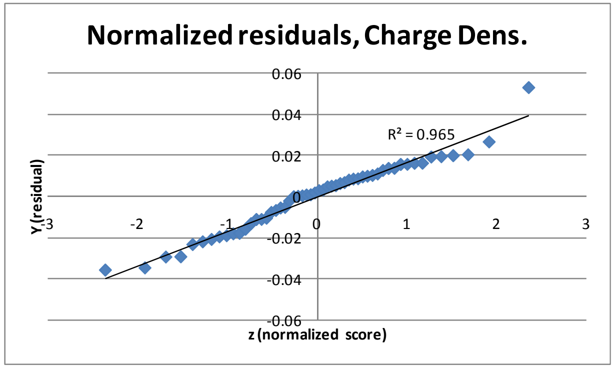
BS_{ik}=Blend-Strain interaction

RS_{jk}=Shear Rate-Strain Interaction

ε_{ijk} = Residual Error



Comparison between predicted and observed values for charge density. The factors blend, shear rate, and strain, and their two-way interactions account for 95% of all the variability in the data set.

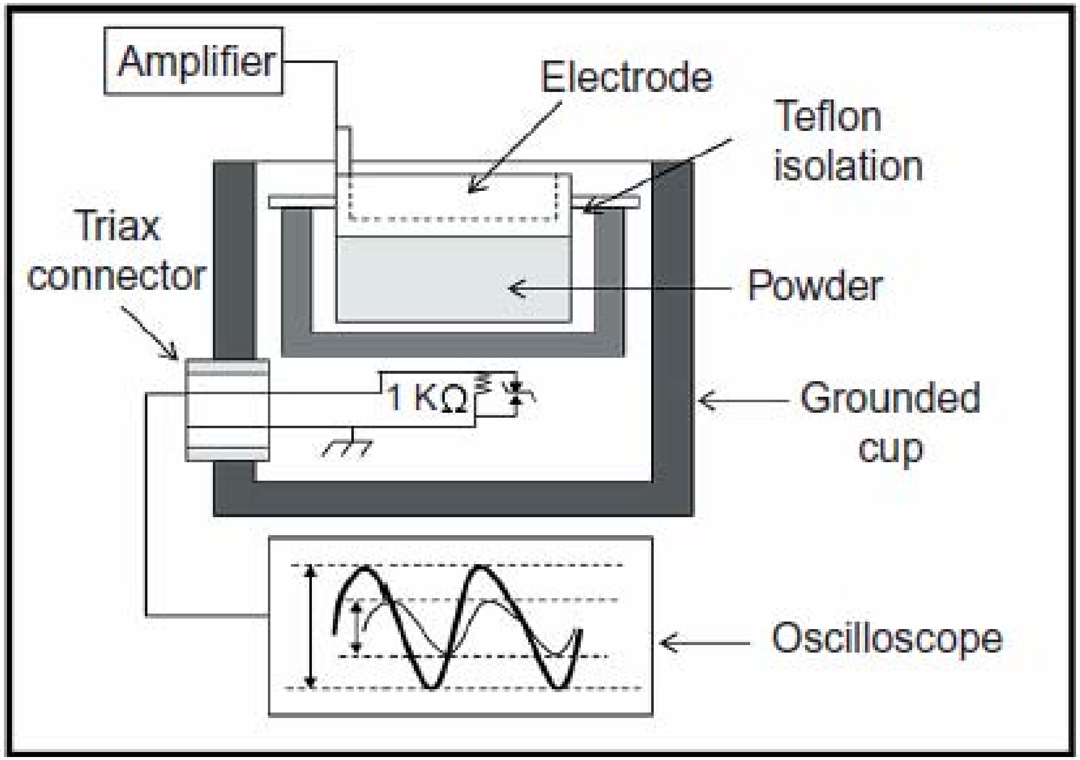


Test of normality for residuals of the observed charge density measurements. Clearly, the residuals are normally distributed.

Main ANOVA Charge Density

Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
blend	0.23118	5	0.046236	60.15405	2.30768E-11	2.710889837
shear rate	0.006637	2	0.003319	4.317552	0.027625633	3.492828477
strain	0.001629	2	0.000815	1.059872	0.365173327	3.492828477
blend*shear rate	0.053338	10	0.005334	6.939399	0.000127081	2.347877567
Blend*strain	0.017712	10	0.001771	2.304363	0.053754257	2.347877567
shear rate*strain	0.004212	4	0.001053	1.369861	0.279759419	2.866081402
Error	0.015373	20	0.000769			
Total	0.330081	53				

Methods: Impedance



Pingali et al (2009)

- *Voltage is supplied to Faraday cup loaded with 40g sample*
- *Function generator and TREK amplifier generate output high voltage*
- *Peak-to-peak voltage at several applied frequencies is recorded*

$$R_p = \frac{V_s}{I_{out}}$$

- *Impedance is recorded as ratio of supplied voltage and output current*
- *Impedance value at lowest measured frequency (100Hz) is taken as an index value for comparison to flow measurements*

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Effect of Shear Rate, Strain, and Blend Composition on Electric Properties

Impedance

No MgSt

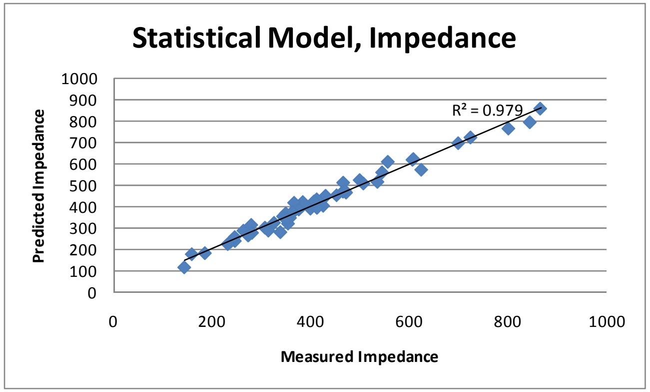
With MgSt

Effect of Shear Rate, Strain, and Blend Composition on Electric Properties

Impedance

$$Y_{ijk} = \mu + B_i + R_j + BR_{ij} + S_k + BS_{ik} + RS_{jk} + \varepsilon_{ijk}$$

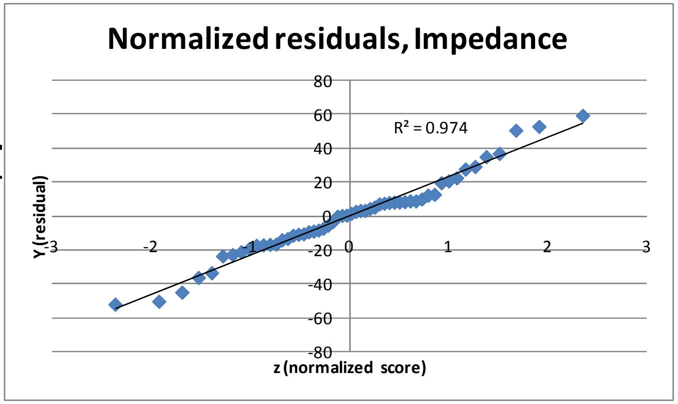
B_i=Blend effect
R_j=Shear Rate effect
BR_{ij}=Blend-Shear Rate interaction
S_k=Strain Effect
BS_{ik}=Blend-Strain interaction
RS_{jk}=Shear Rate-Strain Interaction
ε_{ijk} = Residual Error



Comparison between predicted and observed values for impedance. The factors blend, shear rate, and strain, and their two-way interactions account for 98% of all the variability in the data set.

Main ANOVA Impedance

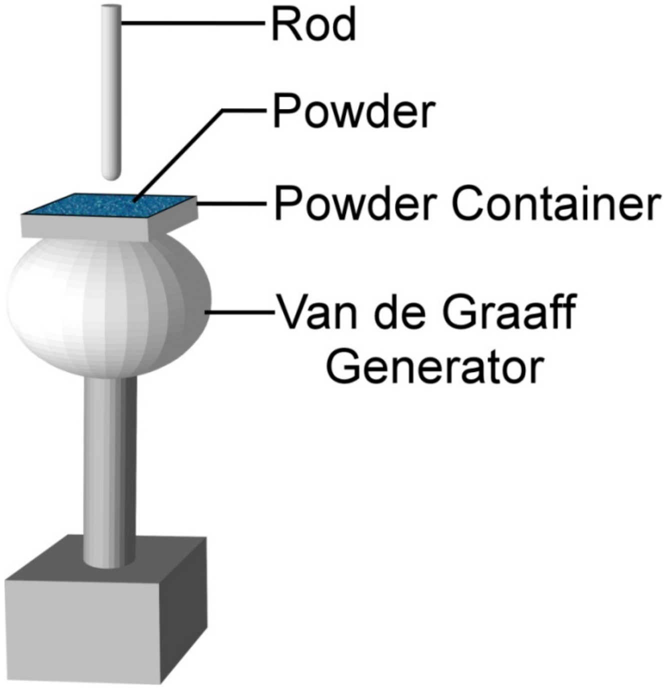
Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
blend	532243.2	5	106448.6	72.63467	3.95828E-12	2.710889837
shear rate	13574.93	2	6787.466	4.631391	0.022240458	3.492828477
strain	18228.16	2	9114.082	6.218946	0.007939147	3.492828477
blend*shear rate	208431.4	10	20843.14	14.22221	5.0522E-07	2.347877567
Blend*strain	599390.2	10	59939.02	40.89907	4.08381E-11	2.347877567
shear rate*strain	5610.956	4	1402.739	0.957152	0.452281797	2.866081402
Error	29310.7	20	1465.535			
Total	1406790	53				



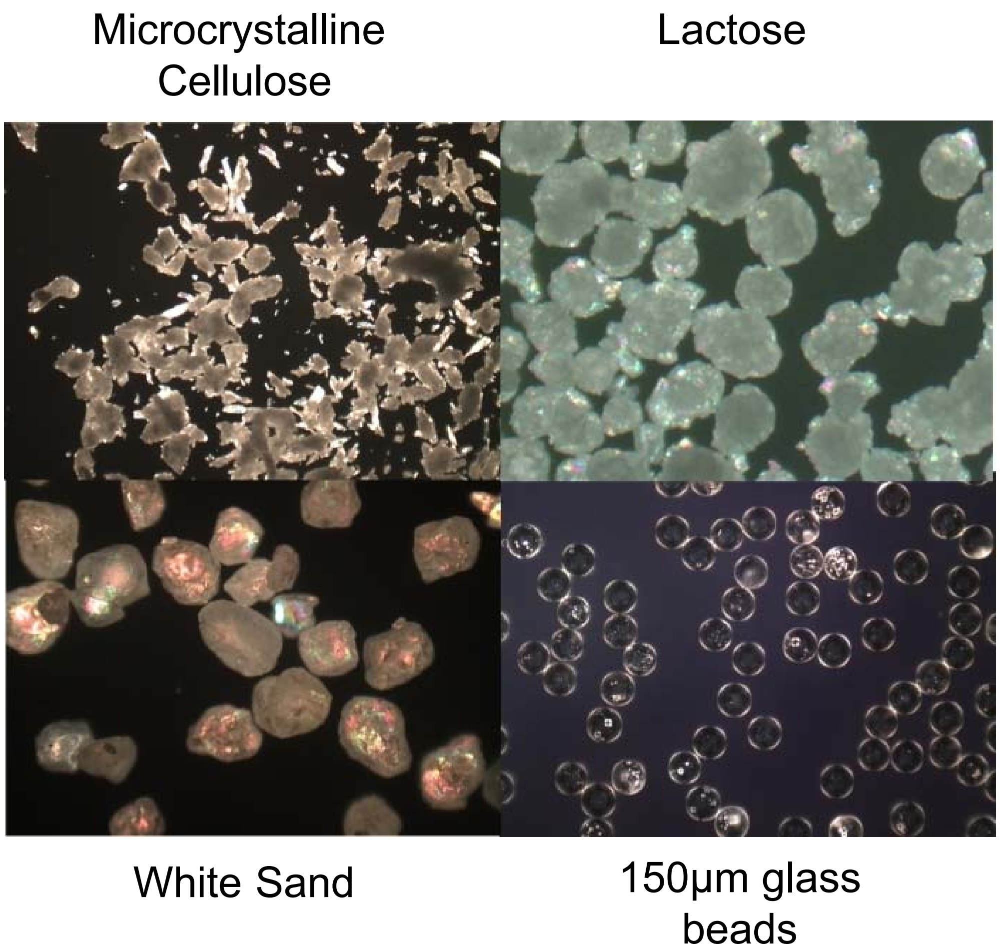
Test of normality for residuals of the observed impedance measurements. The residuals are normally distributed, displaying a R^2 of .97 when compared to a normal distribution.

Powder Adhesion in nonuniform Electric Fields

- Experiments
  - *High voltage produced with Van de Graaff generator*

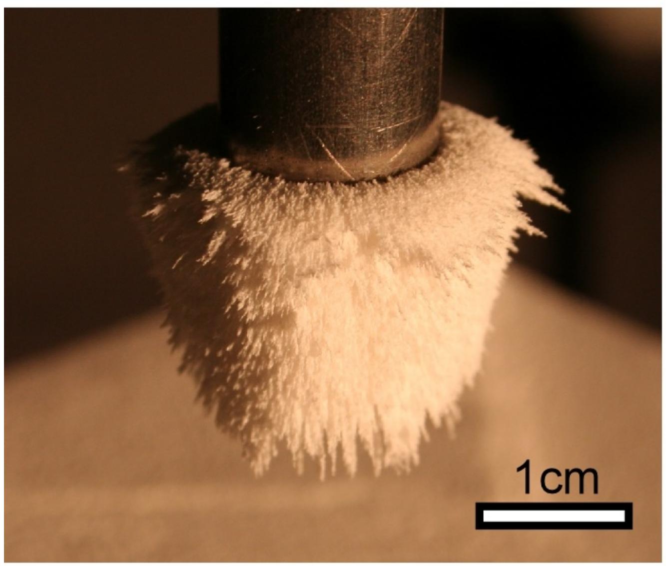


Marche et al., Electrostatic instabilities, charging and agglomeration in flowing granular materials, 2008.

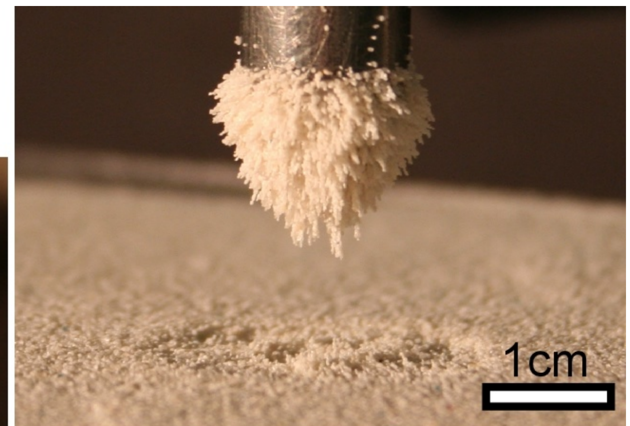


Adhesion

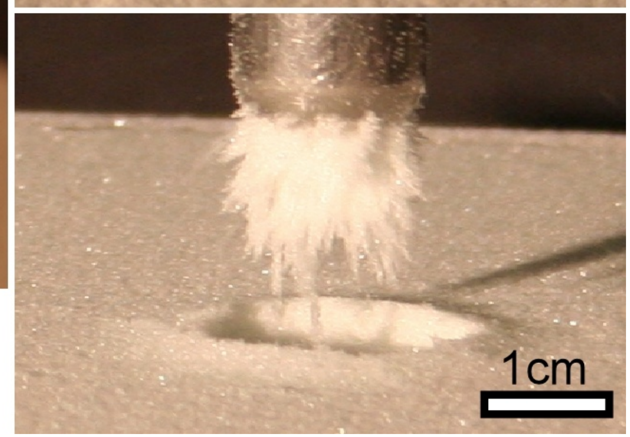
- Materials adhere to a grounded rod in E-field



Cellulose

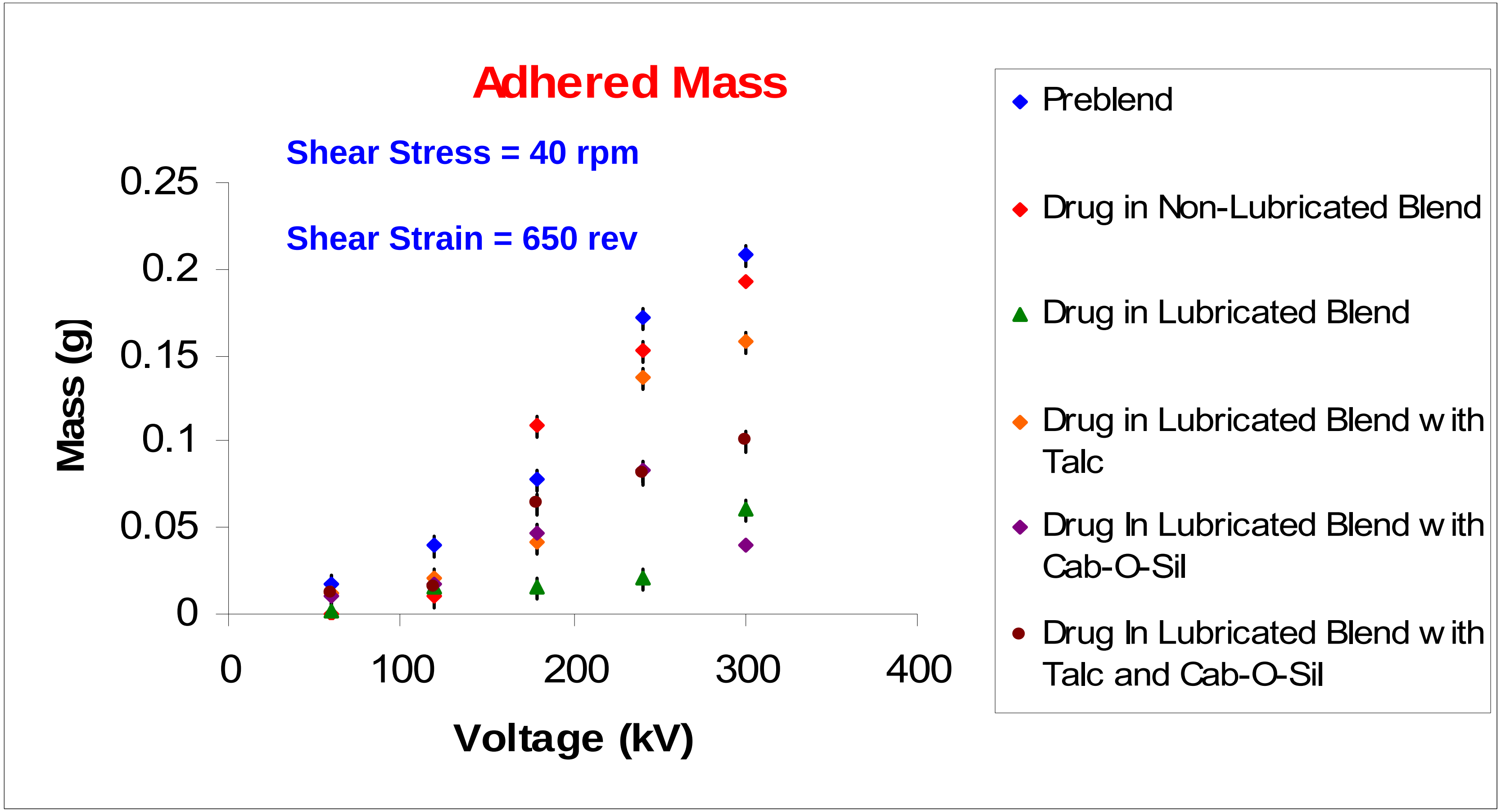


White Sand



Glass Beads

Adhesion and E-field Strength



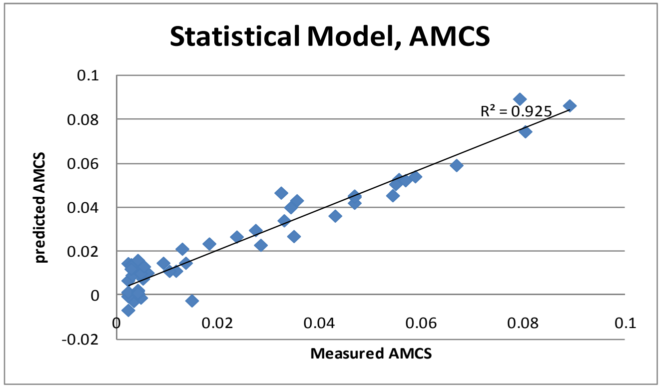
MgSt, Talc and Cab-O-Sil are effective in decreasing adhered mass.

Effect of Shear Rate, Strain, and Blend Composition on Electric Properties

Dielectrophoresis

$$Y_{ijk} = \mu + B_i + R_j + BR_{ij} + S_k + BS_{ik} + RS_{jk} + \varepsilon_{ijk}$$

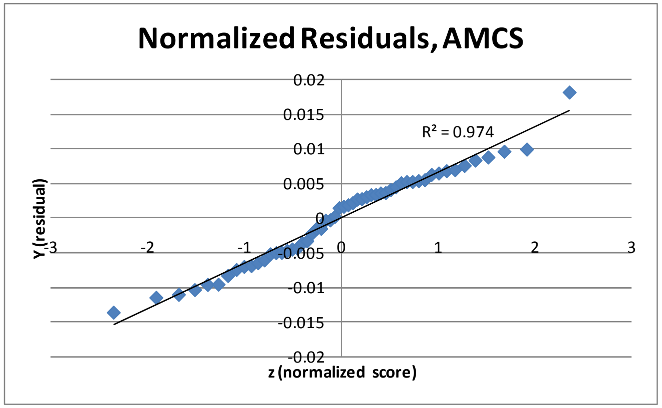
B_i=Blend effect
R_j=Shear Rate effect
BR_{ij}=Blend-Shear Rate interaction
S_k=Strain Effect
BS_{ik}=Blend-Strain interaction
RS_{jk}=Shear Rate-Strain Interaction
ε_{ijk} = Residual Error



Comparison between predicted and observed values for AMCS. The factors blend, shear rate, and strain, and their two-way interactions account for 92% of all the variability in the data set.

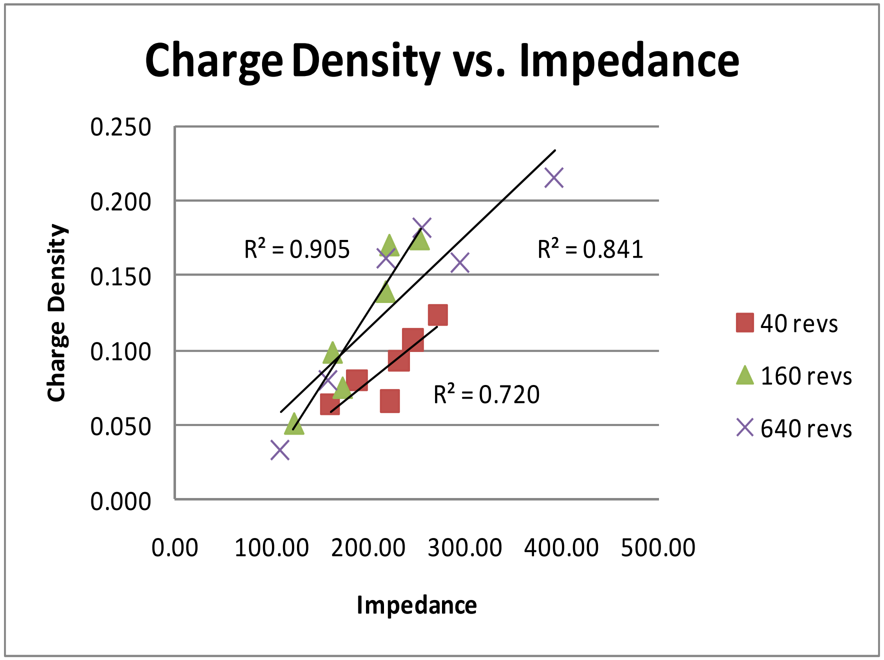
Main ANOVA Adhered mass

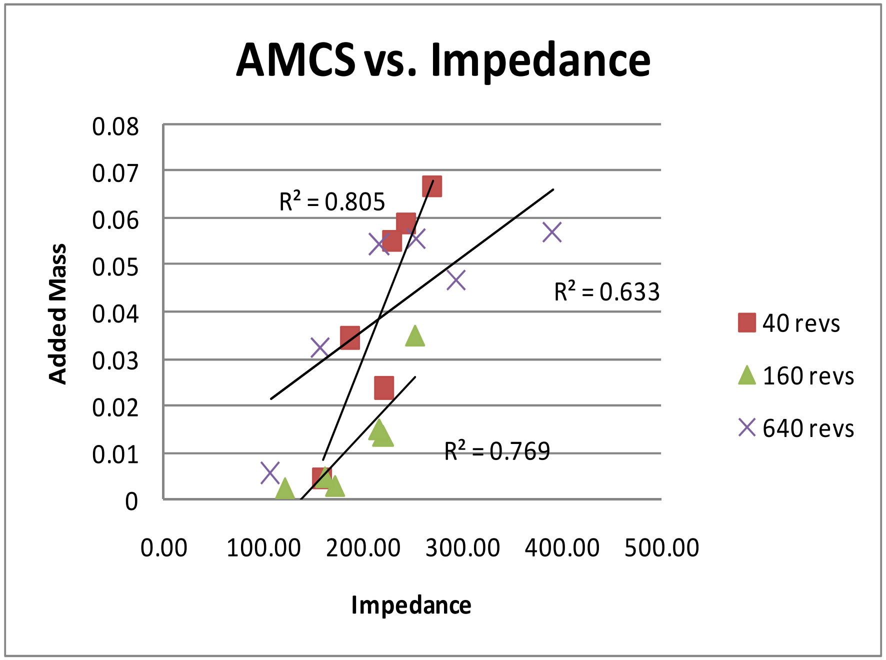
Source of Variation	*SS*	*df*	*MS*	*F*	*P-value*	*F crit*
blend	0.009071	5	0.001814	15.5158	2.72462E-06	2.710889837
shear rate	0.002453	2	0.001227	10.48975	0.000766715	3.492828477
strain	0.001277	2	0.000639	5.461498	0.012808091	3.492828477
blend*shear rate	0.012198	10	0.00122	10.43165	6.18598E-06	2.347877567
Blend*strain	0.001693	10	0.000169	1.447984	0.230378437	2.347877567
shear rate*strain	0.002304	4	0.000576	4.925993	0.006271982	2.866081402
Error	0.002339	20	0.000117			
Total	0.031335	53				



Test of normality for residuals of the observed impedance measurements. The residuals are normally distributed, displaying a R^2 of .97 when compared to a normal distribution.

Effect of Strain and Composition on Electric Properties

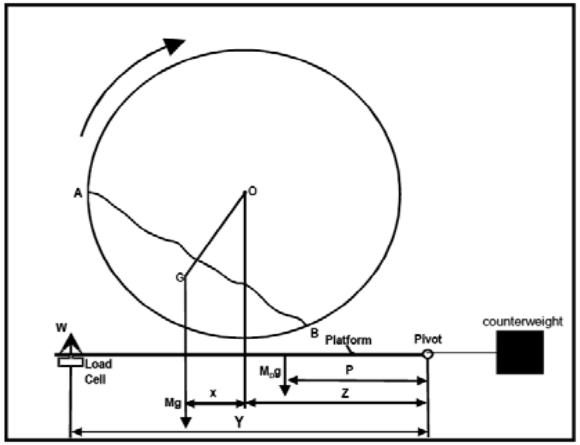


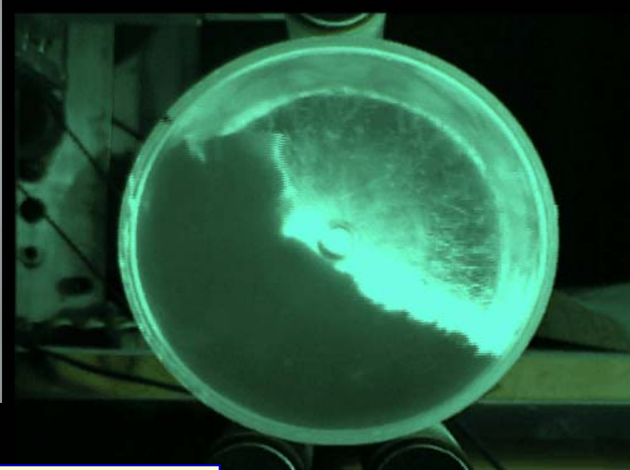


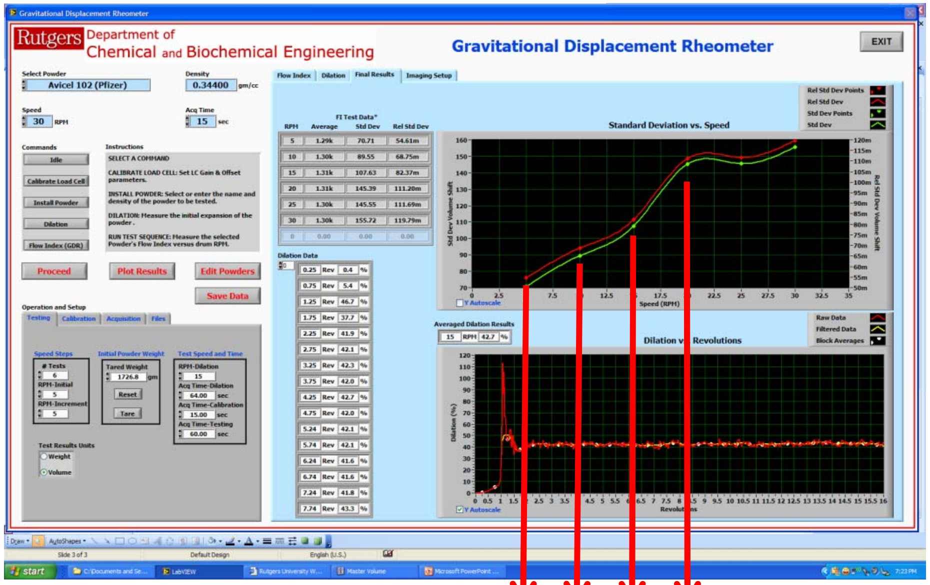
Correlation of Charge Density vs. Impedance. A high degree of correlation between charge density and impedance is observed across blends and strain levels.

Correlation of AMCS vs. Impedance for the samples in Table (8). A high degree of correlation between dielectrophoresis and impedance is observed across blends and strain levels.

Flow Measurement of Powders



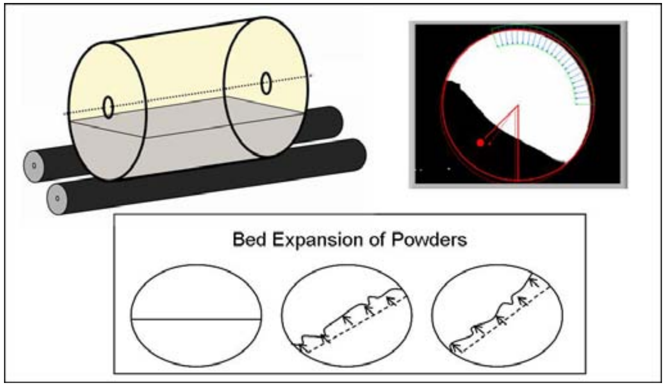


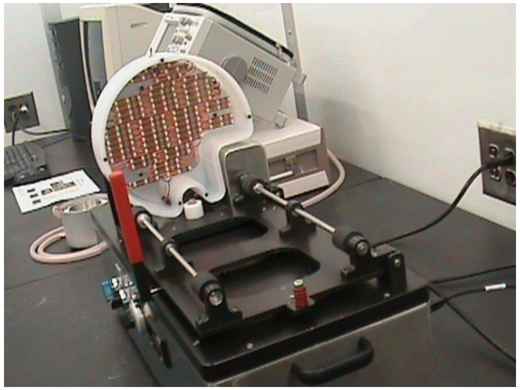


Flow Index

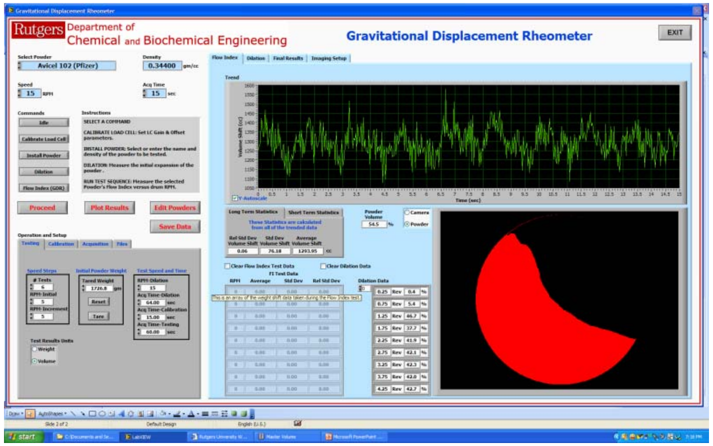
This program displays the changes (standard deviations) in center of mass/volume over a range of rotation rates. The protocol for powder testing includes calibration of load cell (to ensure uniformity of measurement and testing using the standard drum), selecting the powder of known bulk density, imaging set up and running test sequence. The step wise prompts guides the user to carry out a sequence of testing procedure featuring automation of measuring technique of load cell data acquisition, automatic data retrieval, report generation and printout.

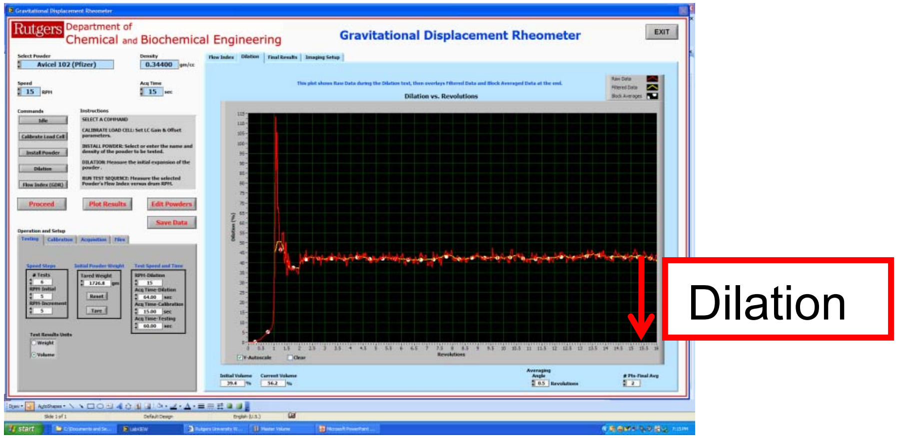
Dilation Measurement





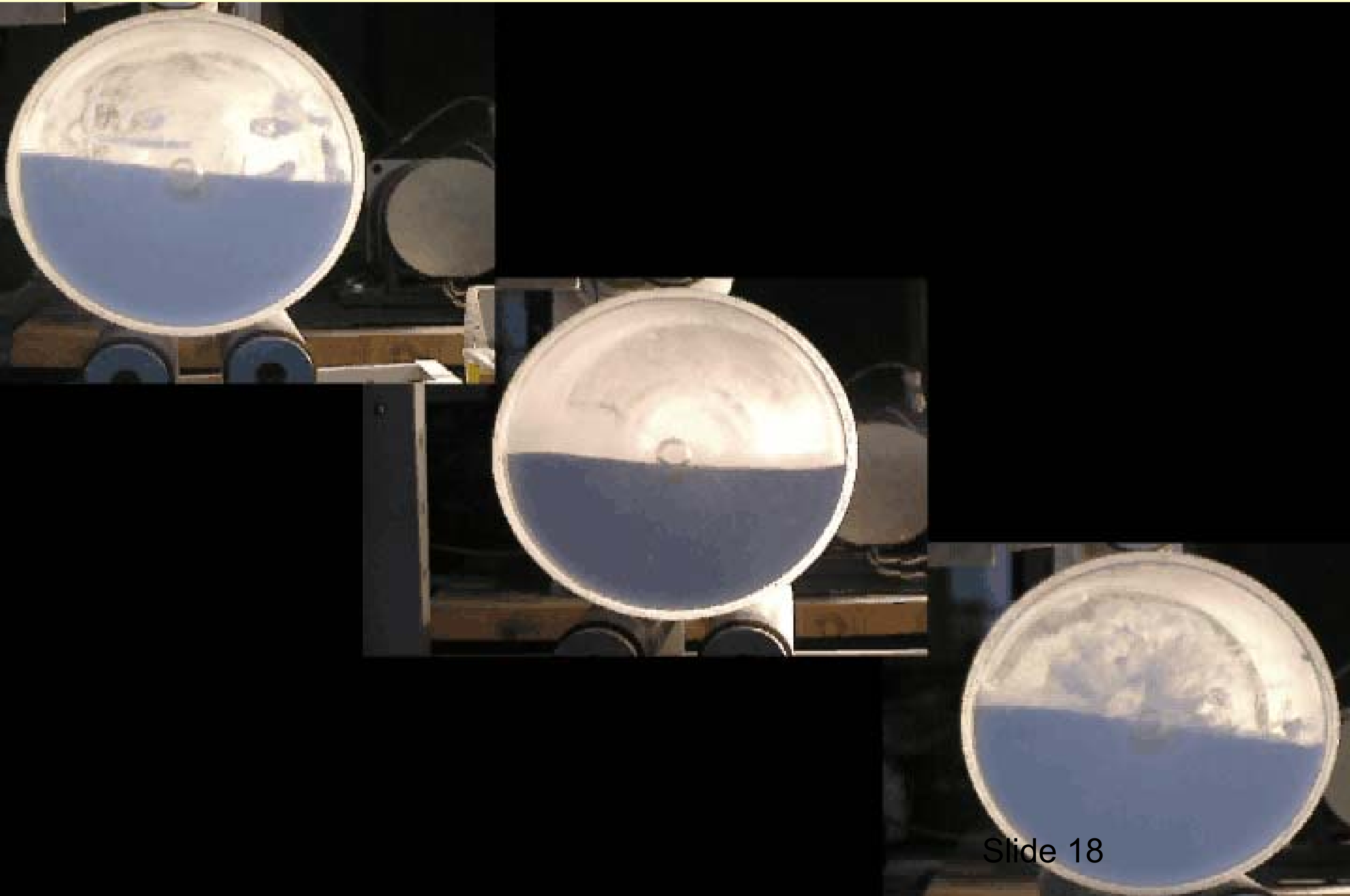
Dilation assembly consists of LED backlighted GDR drum with fire wire camera (640 x 480 pixels) causing the shadow on the end of the drum.



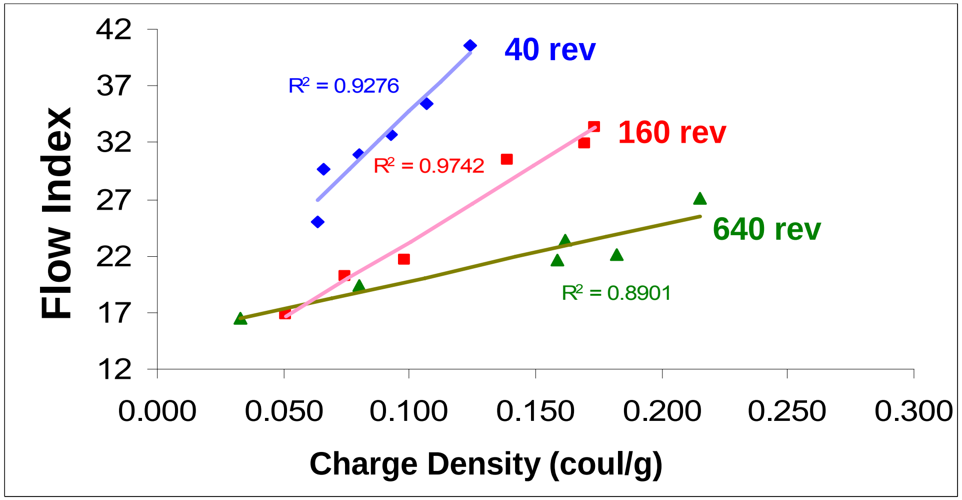


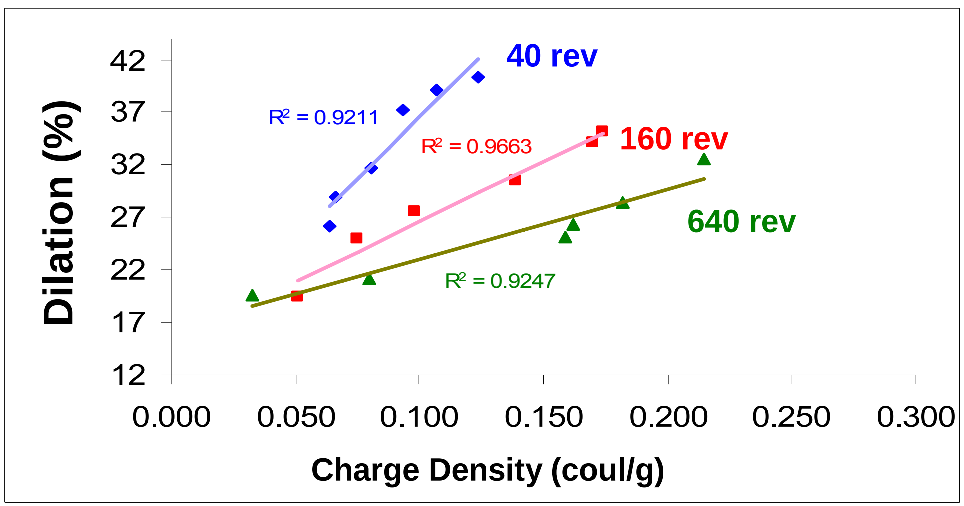
As the drum rotates, the geometric shift of the powder as a percentage of drum radius is displayed continuously.

Dilation needed to flow



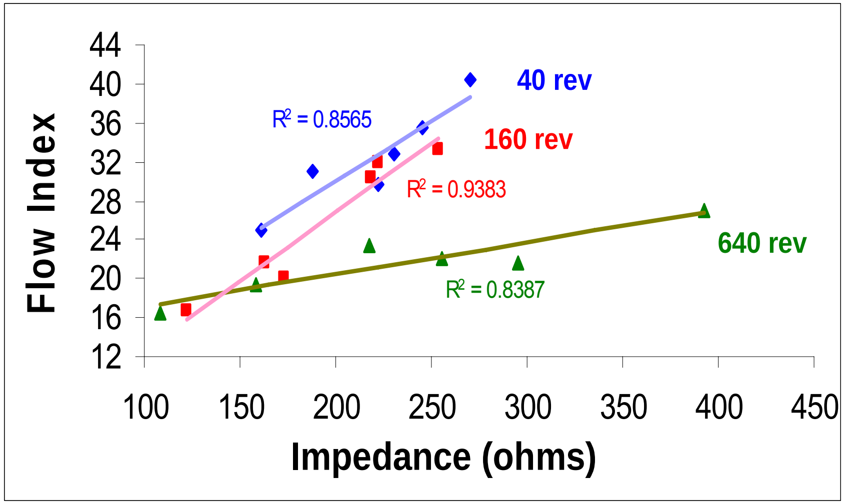
Effect of composition and strain on flow properties: an electric connection

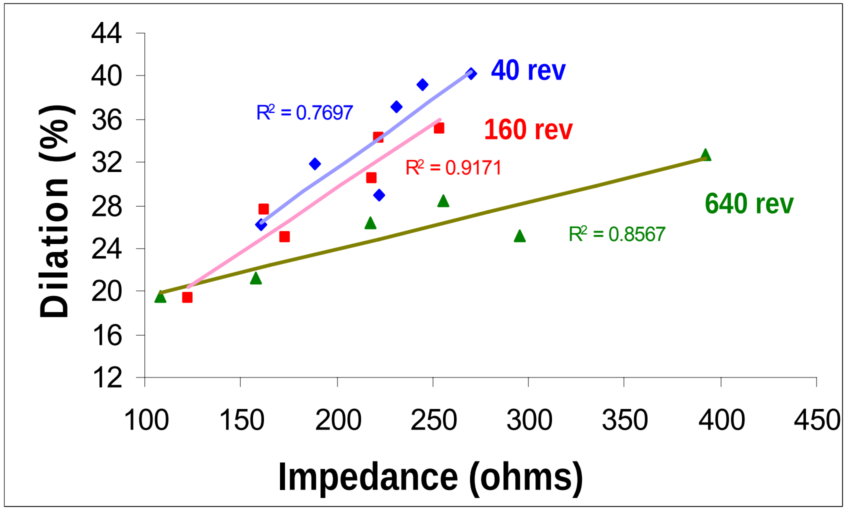




Flow index and dilation correlate to charge acquisition for different shear treatments. Flow index and dilation increased with charge acquisition indicating worsening of powder flow with charge accumulation.

Effect of composition and strain on flow properties: an electric connection





(a) Flow index and (b) dilation correlate to impedance for different strain treatments. A decrease in cohesion is observed with an increase in strain.

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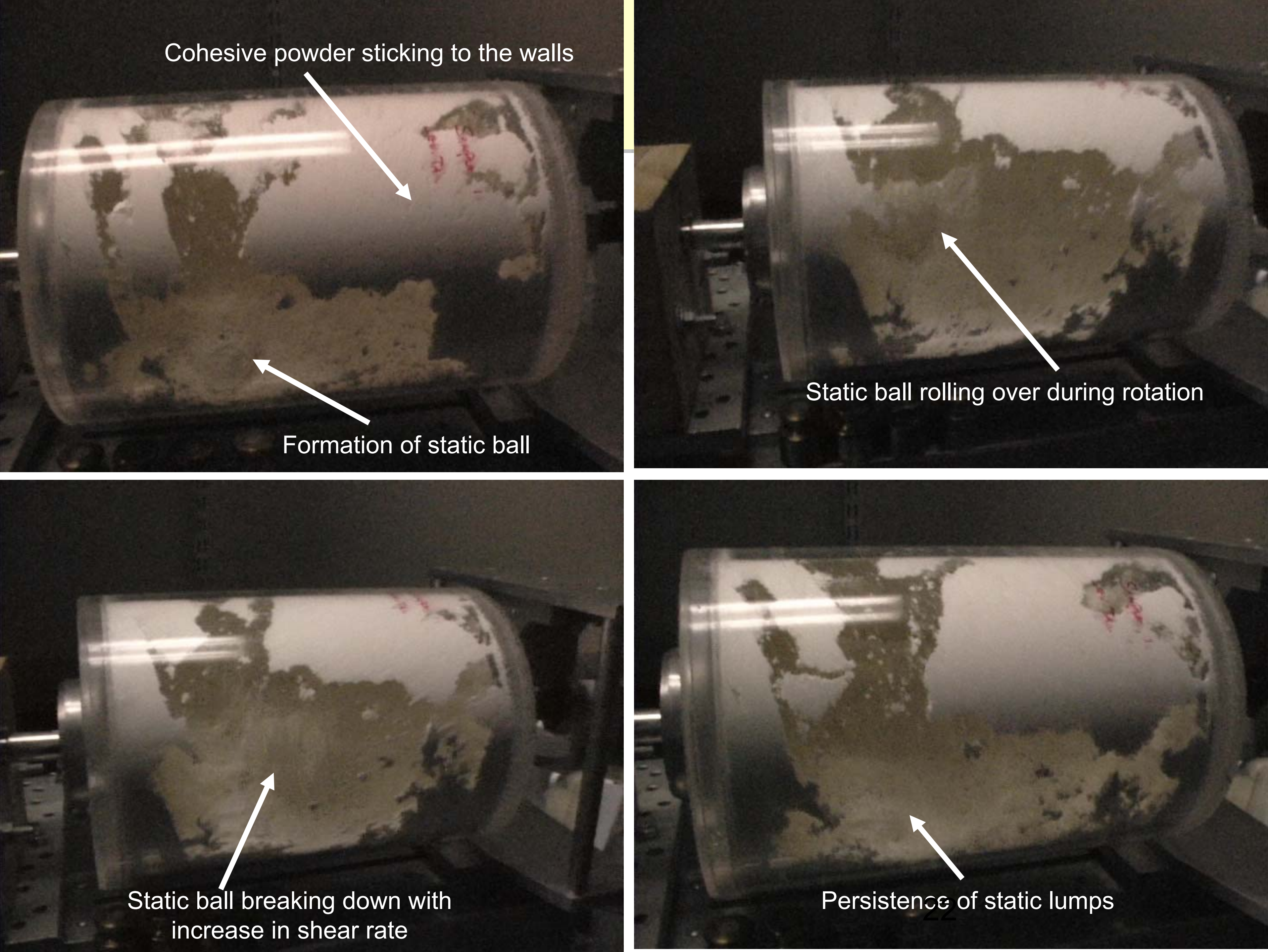
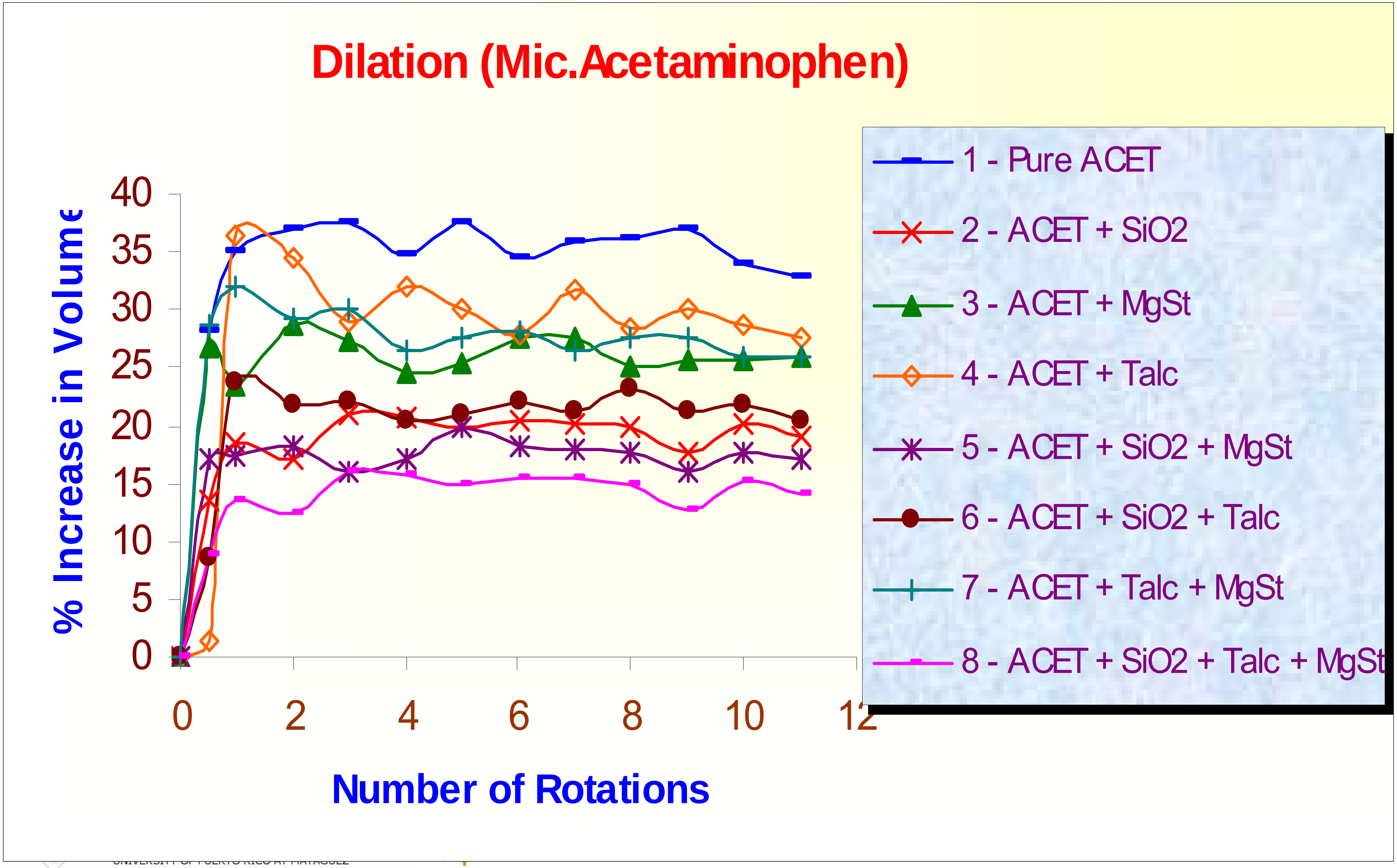
Case Study 2: APIs

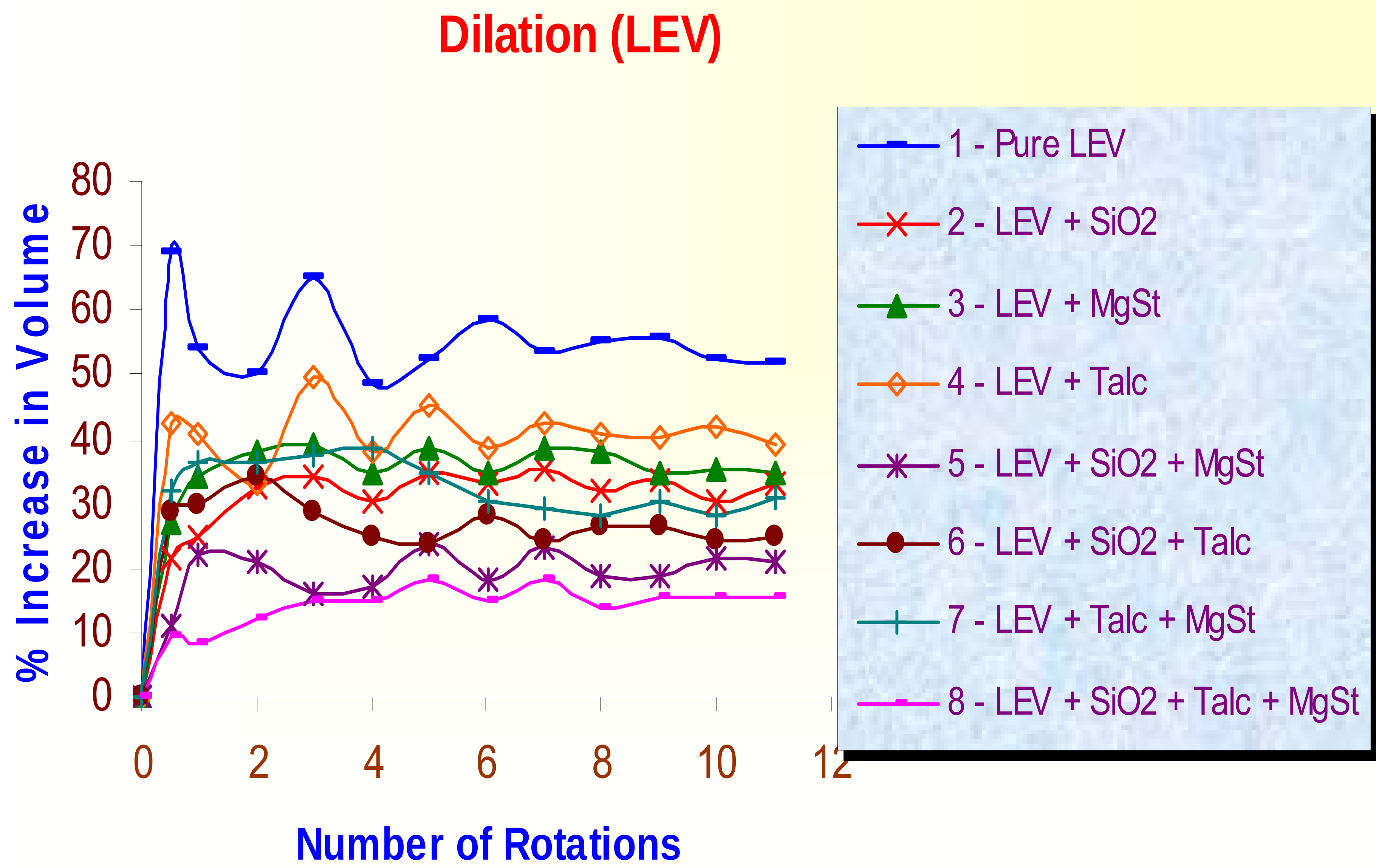
- Acetaminophen
- LEV
- DDMS

- SiO2, MgSt, Talc
- High shear blending in a V-blender with IB
- Flow index, dilation, impedance

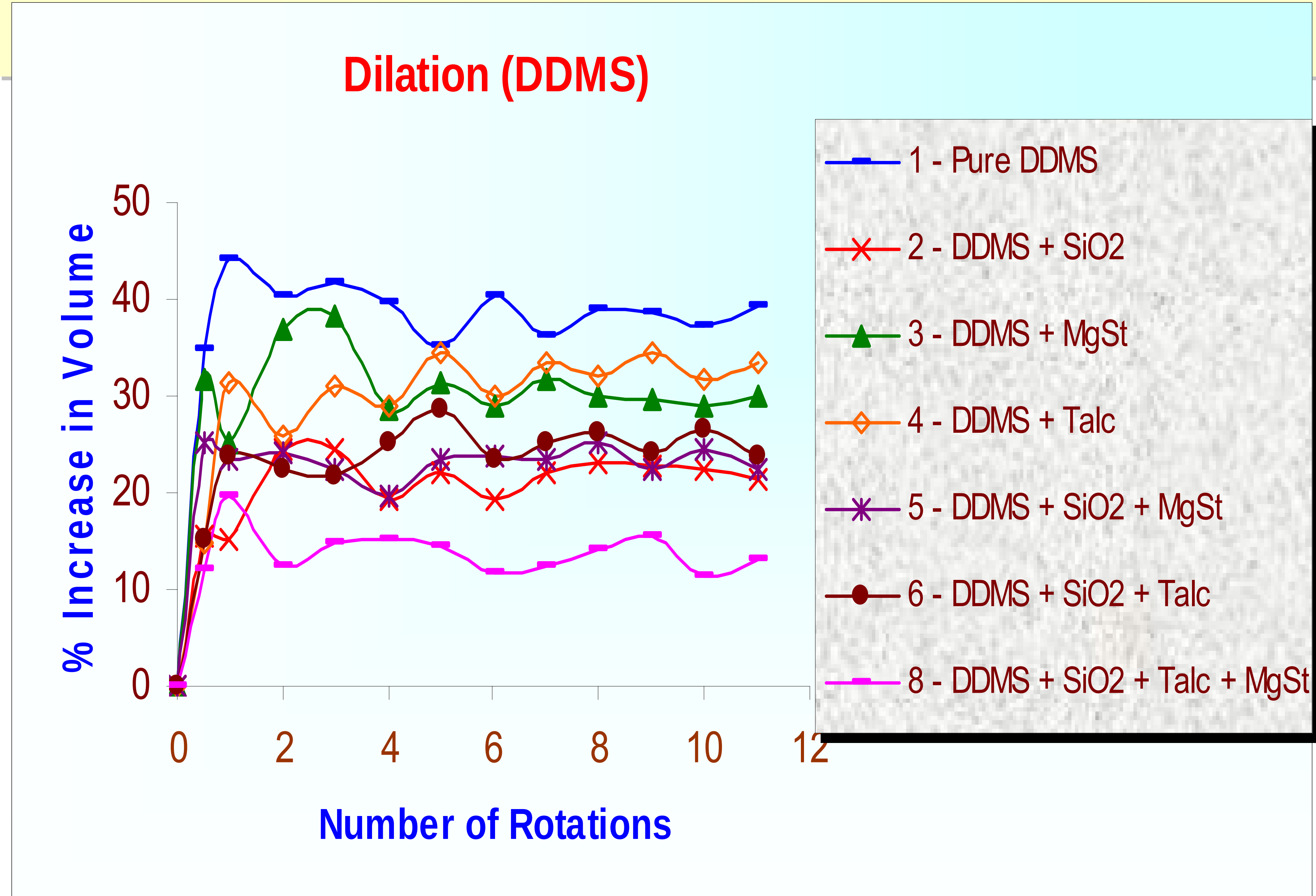

Cohesive powder sticking to the walls
Formation of static ball
Static ball rolling over during rotation
Static ball breaking down with increase in shear rate
Persistence of static lumps

Flow Properties of Acetaminophen Blends

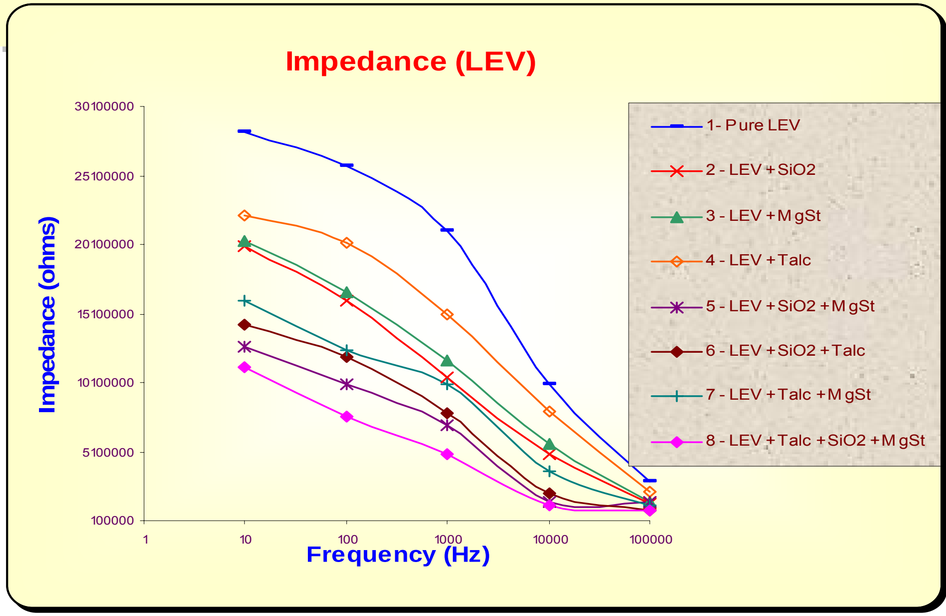


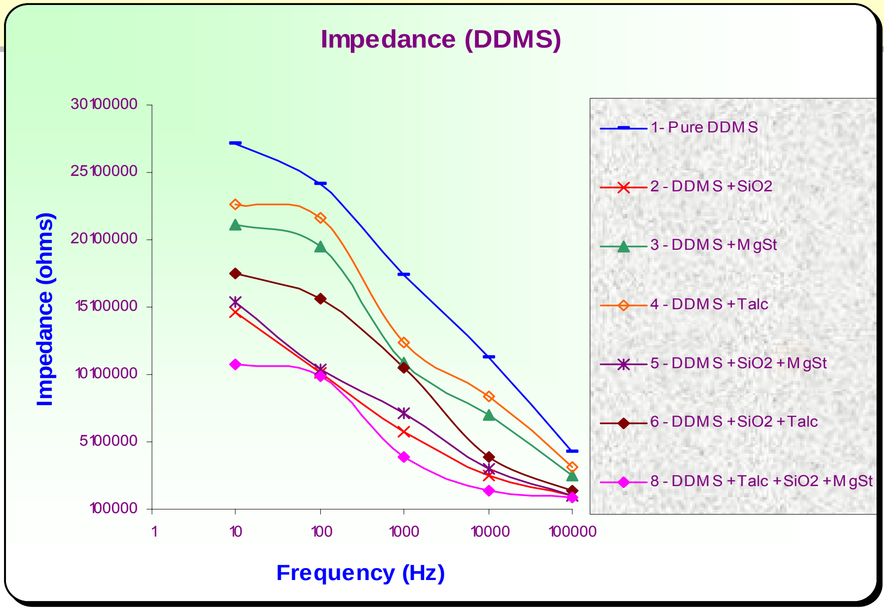


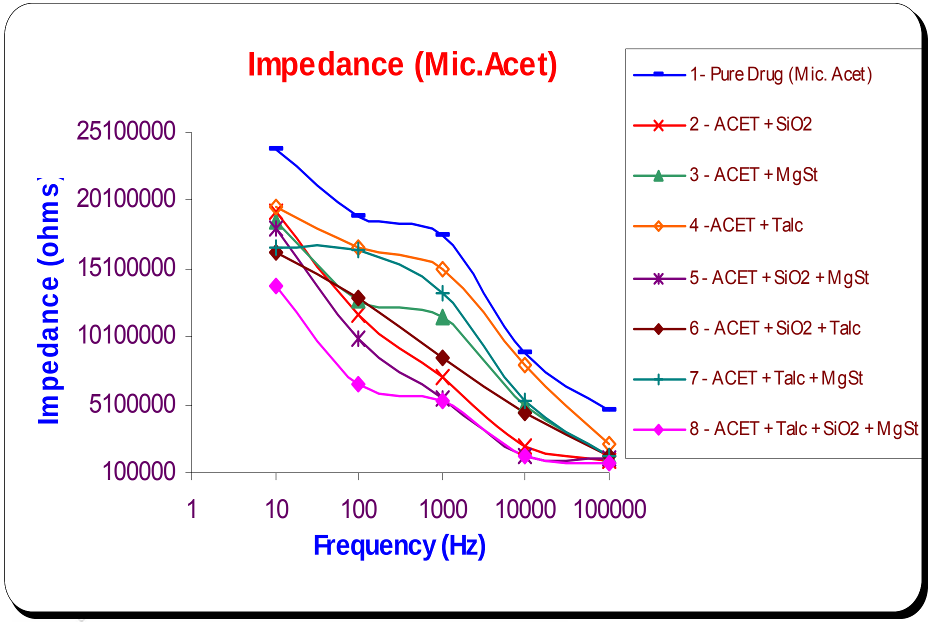
Dilation (LEV)
% Increase in Volume
0
10
20
30
40
50
60
70
80
0
2
4
6
8
10
12
Number of Rotations
1 - Pure LEV
2 - LEV + SiO2
3 - LEV + MgSt
4 - LEV + Talc
5 - LEV + SiO2 + MgSt
6 - LEV + SiO2 + Talc
7 - LEV + Talc + MgSt
8 - LEV + SiO2 + Talc + MgSt


Dilation (DDMS)
% Increase in Volume
0
10
20
30
40
50
0
2
4
6
8
10
12
Number of Rotations
1 - Pure DDMS
2 - DDMS + SiO2
3 - DDMS + MgSt
4 - DDMS + Talc
5 - DDMS + SiO2 + MgSt
6 - DDMS + SiO2 + Talc
8 - DDMS + SiO2 + Talc + MgSt

Impedance

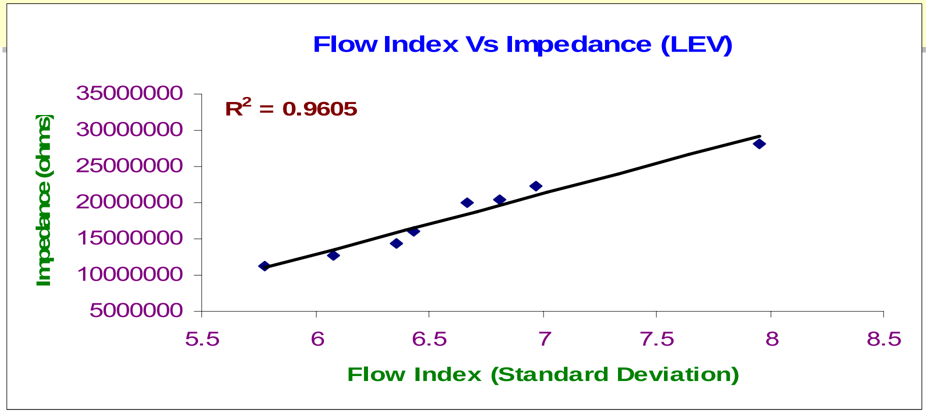


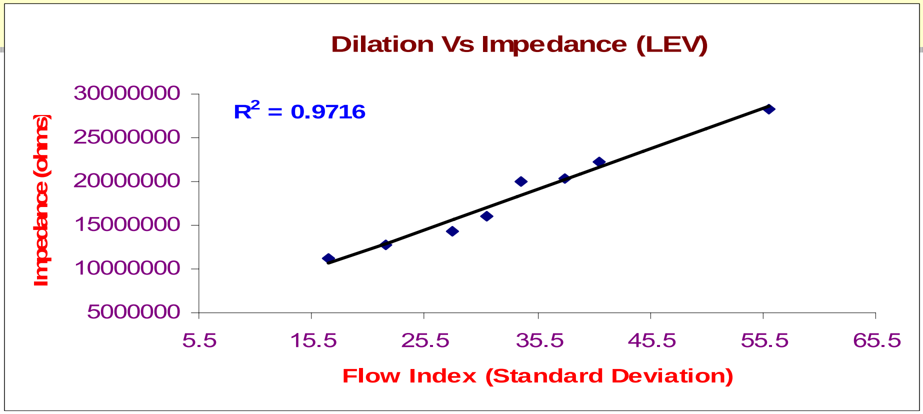


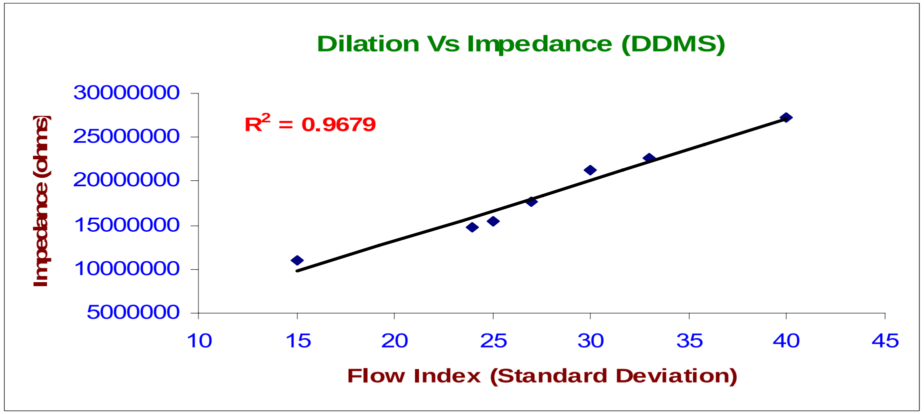


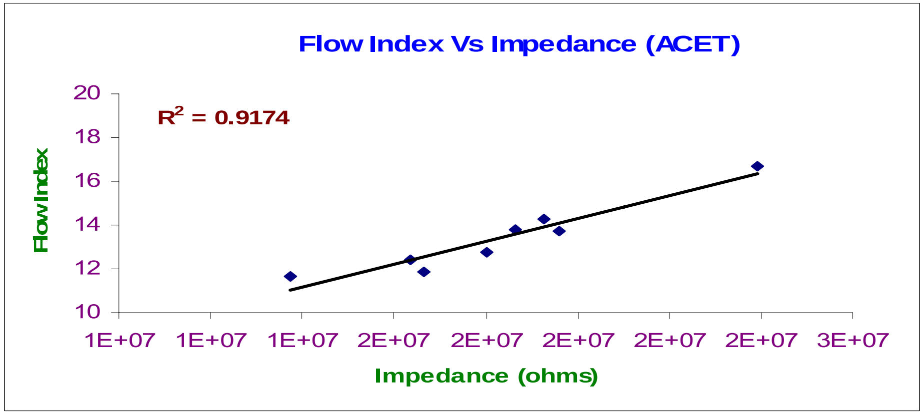
Again, the rank-ordering of impedance results is identical to the rank-ordering of flow index and dilation results of the blends. In all the cases, combination of additives improved the powder flow and decreased impedance.

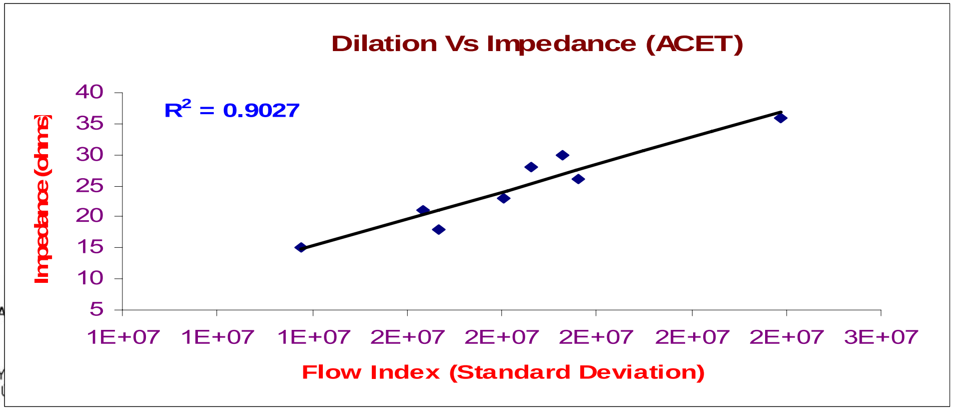
Relation Between Flow And Electrical Properties for APIs





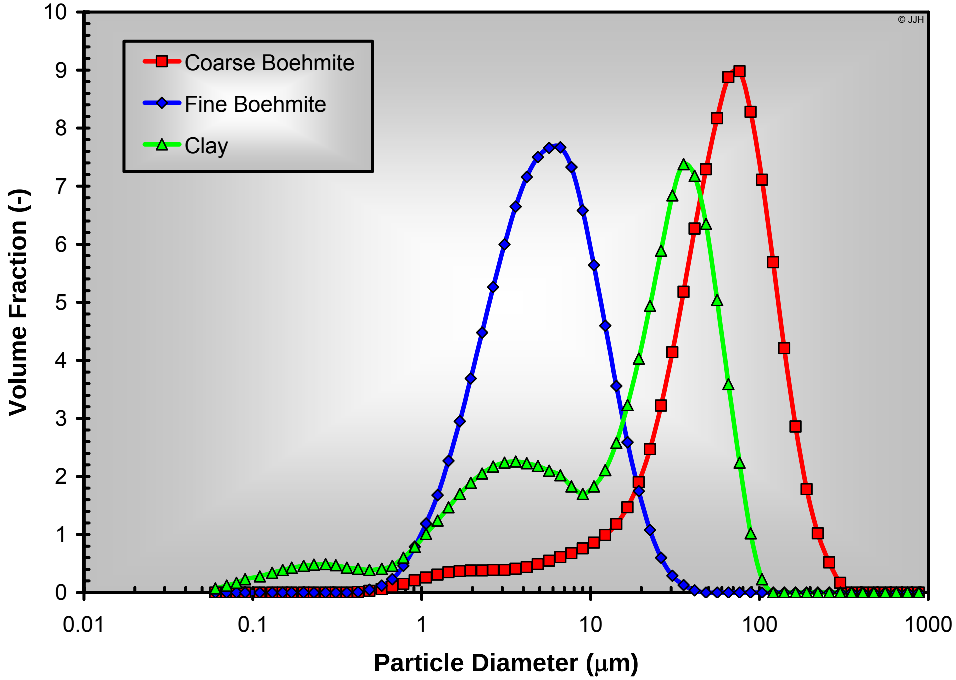






Case study 3: Ceramics

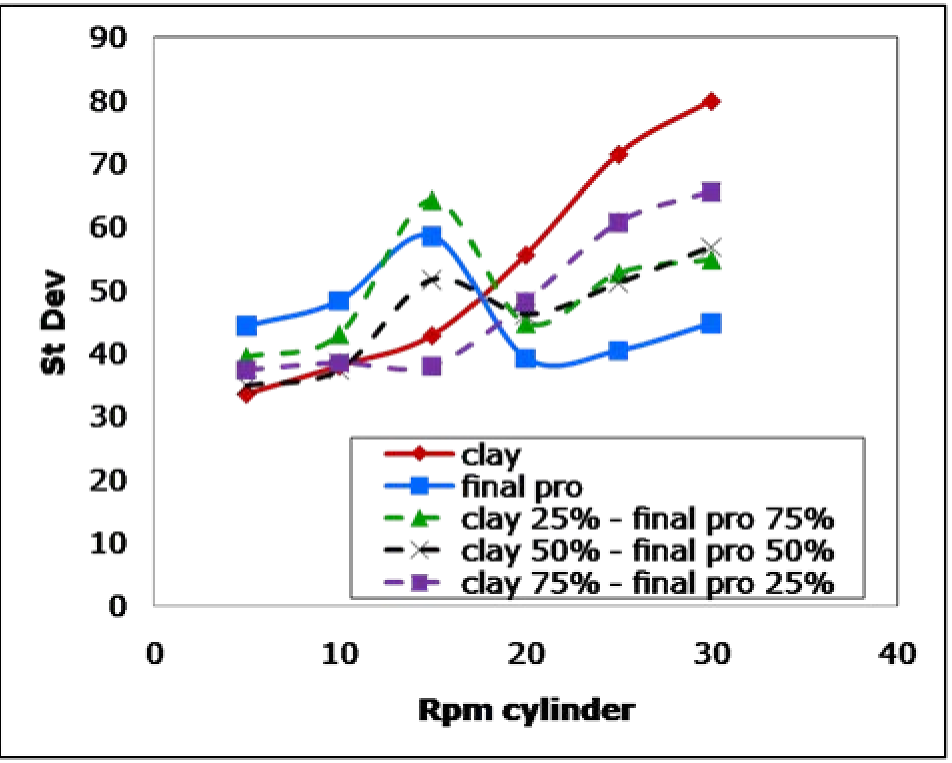
- Three materials:
  - *Fine Boehmite (dp = 5μm) - **FB***
  - *Clay (dp = 22μm) - **C***
  - *Coarse Boehmite (dp = 60μm) - **CB***

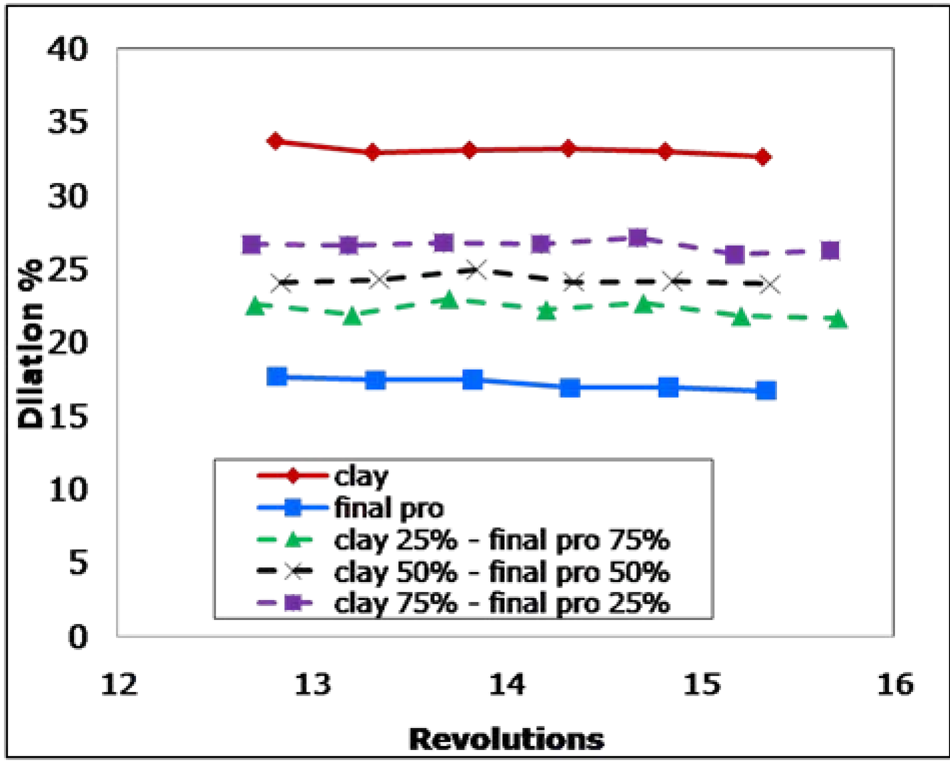


(PSD data courtesy AlbeMarle)

- Blends (ranked from worst to best flow):
  - *100% Fine Boehmite*
  - *75% Fine Boehmite and 25% Clay*
  - *50% Fine Boehmite and 50% Clay*
  - *25% Fine Boehmite and 75% Clay*
  - *100% Clay*
  - *75% Clay and 25% Coarse Boehmite*
  - *50% Clay and 50% Coarse Boehmite*
  - *25% Clay and 75% Coarse Boehmite*
  - *100% Coarse Boehmite*

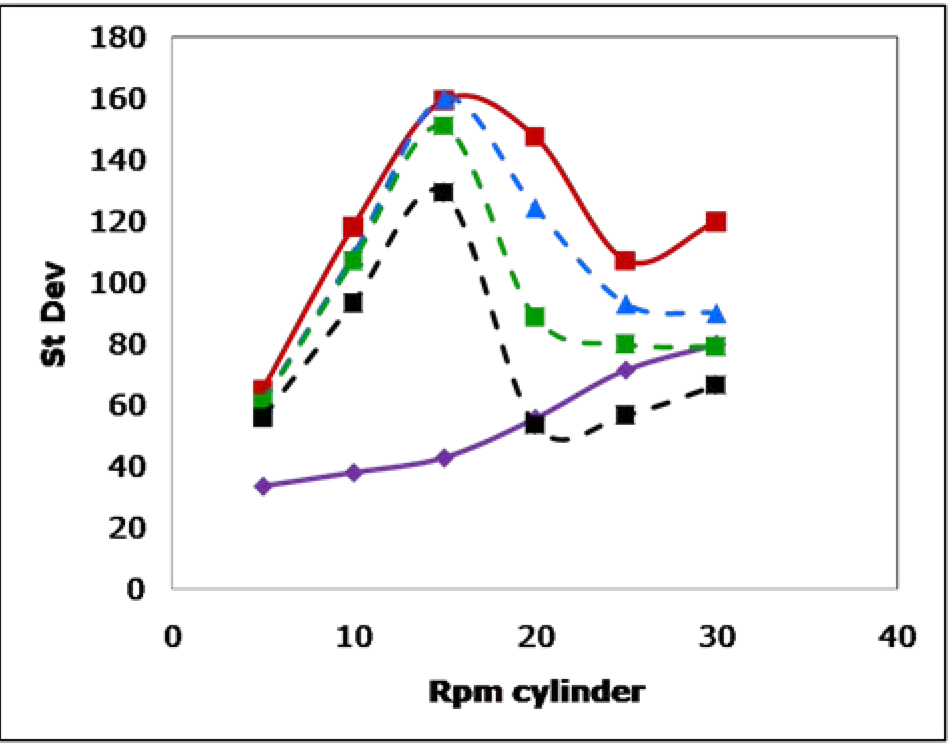
GDR and Dilation Data (intermediate to best flow)

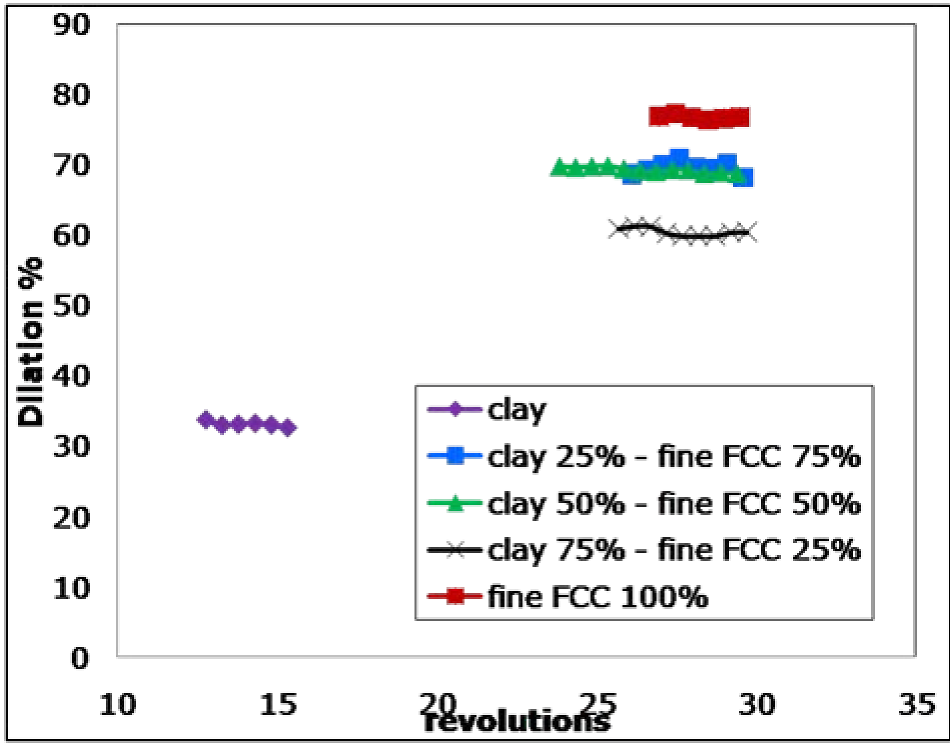




- *The flow ranking was expected to be pure clay (worst) → clay (75%) → clay (50%) → clay (25%) → CB (best)*
- *The flow behavior of mixtures was in direct correlation with the component concentration*

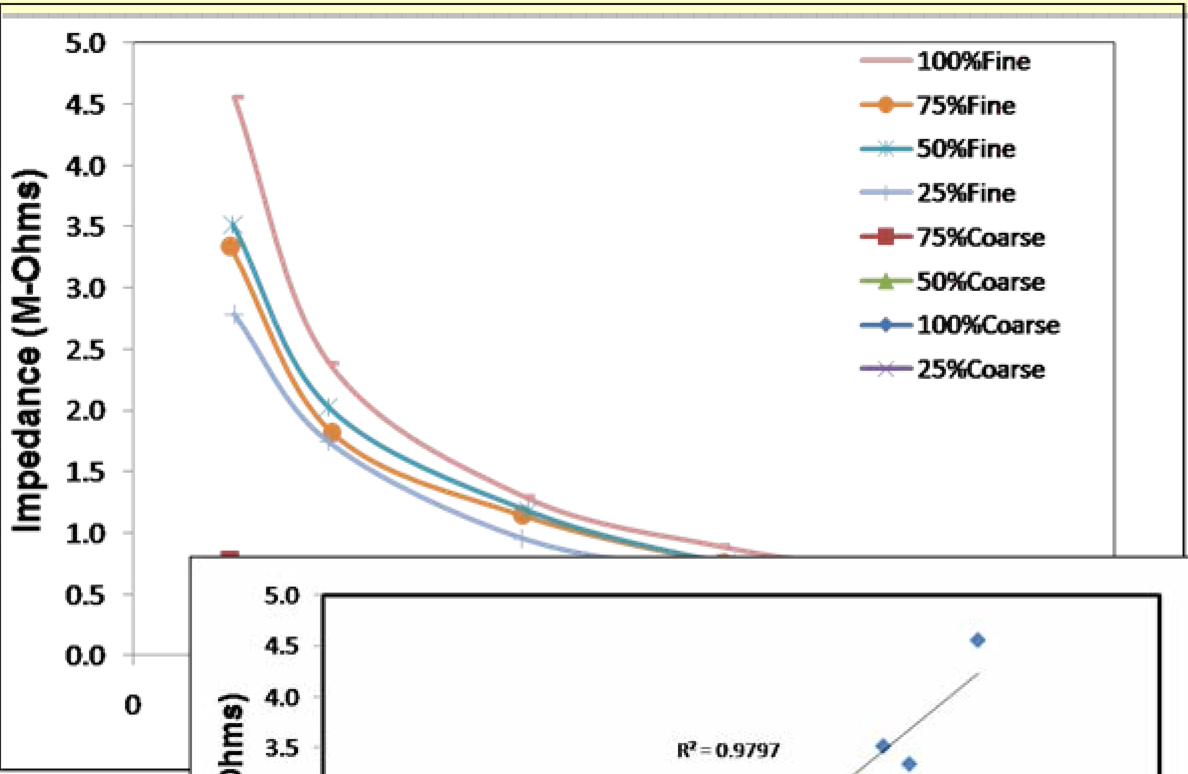
GDR and Dilation Data (worst to intermediate flow)

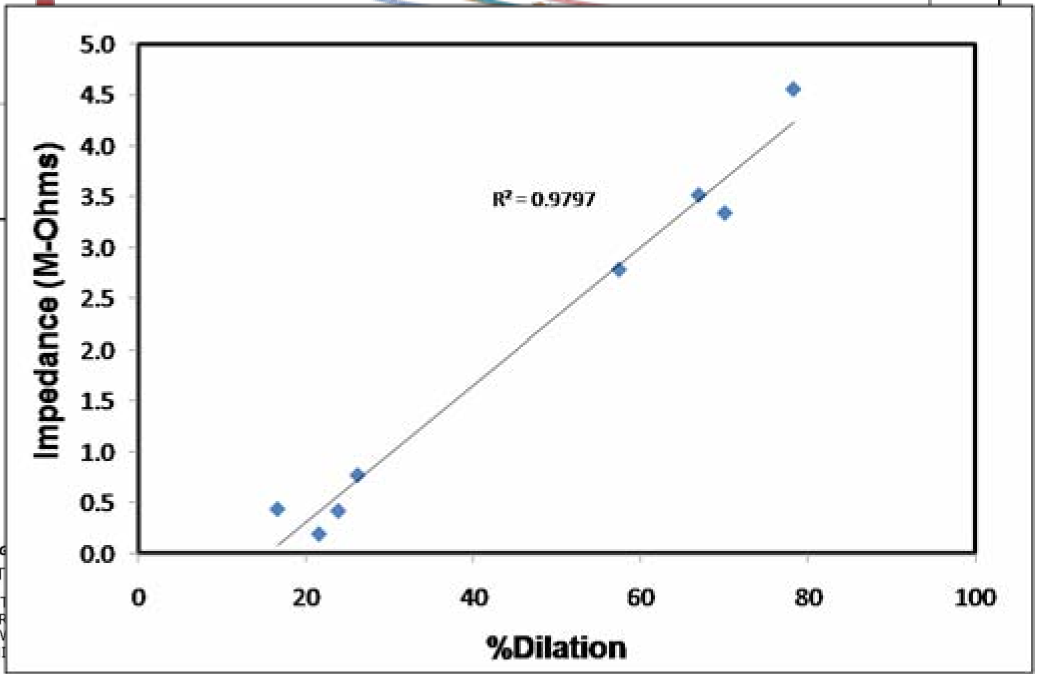




- ***<u>In case of FB/C mixtures</u>**, the flow behavior of the blends was governed by the presence of fine boehmite.*

Impedance





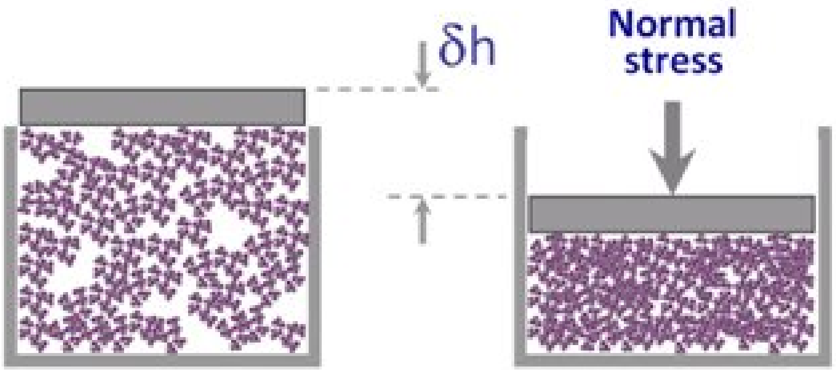
- *Impedance is related to cohesion through*
  1) *Larger capacitance = increased interparticle forces*
  2) *Lower density = lower conductivity = higher resistance*

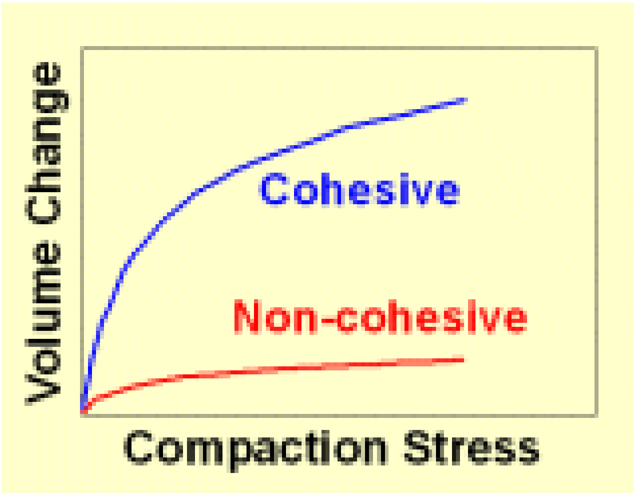
- *Impedance correlates well with dilation measurements, suggesting that it can be used as a flow properties measurement*

Compressibility Test

- Measures cohesion by relating it to the amount of entrapped air
- Technique shows general correlation to particle size and therefore cohesion, but some exceptions exist
- <u>*Procedure*</u>:
  - *Powder is conditioned by a helical blade passing through*
  - *Normal force is subsequently applied with a vented piston in intervals from 0.5kPa to 15kPa*
  - *The change of volume upon compression is measured at each step*
  - *The compressibility index (I_C) is calculated as*

$$I_C = \frac{\rho_{compr}}{\rho_{cond}} \times 100\%$$





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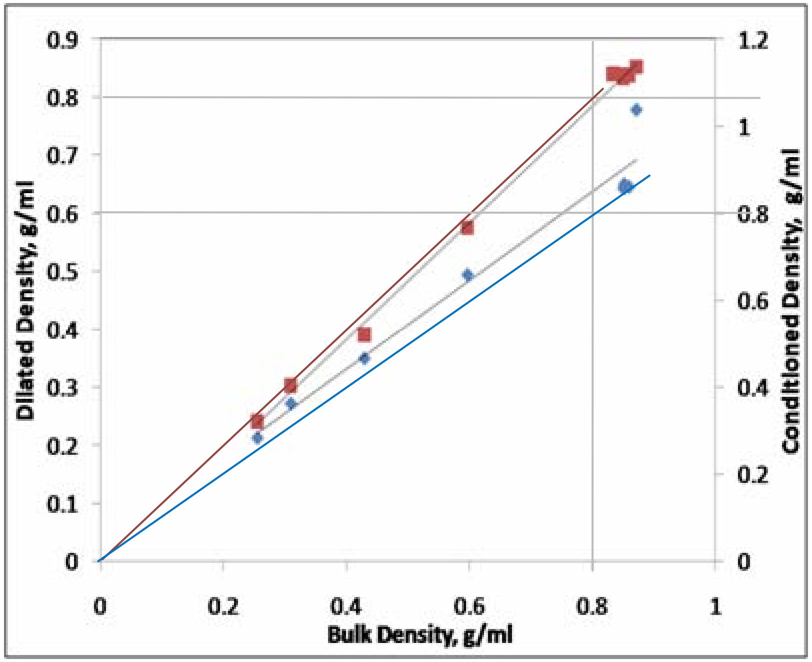
Density Comparisons



- *Both, <u>FT4 Conditioned Density</u> and <u>GDR Dilated Density</u>, correlate remarkably well to the Bulk (Poured) Density measurements*

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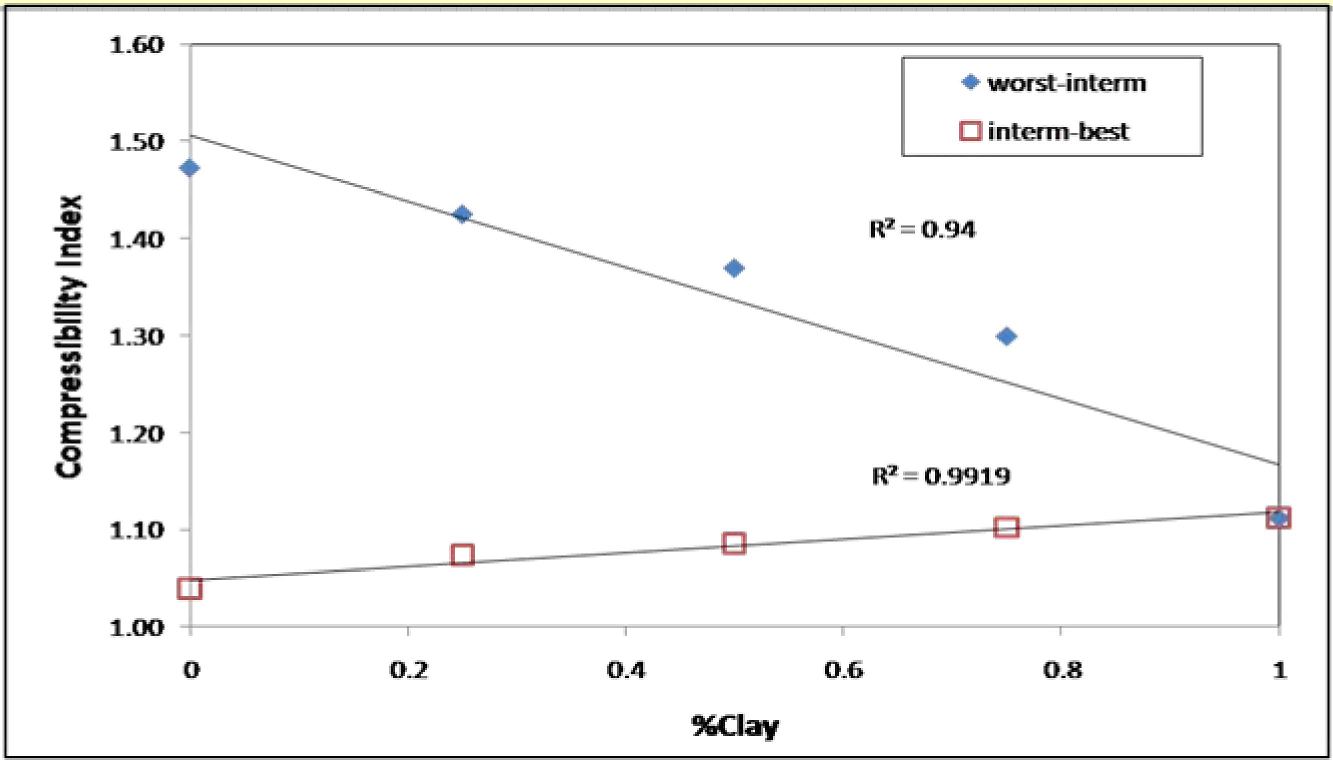
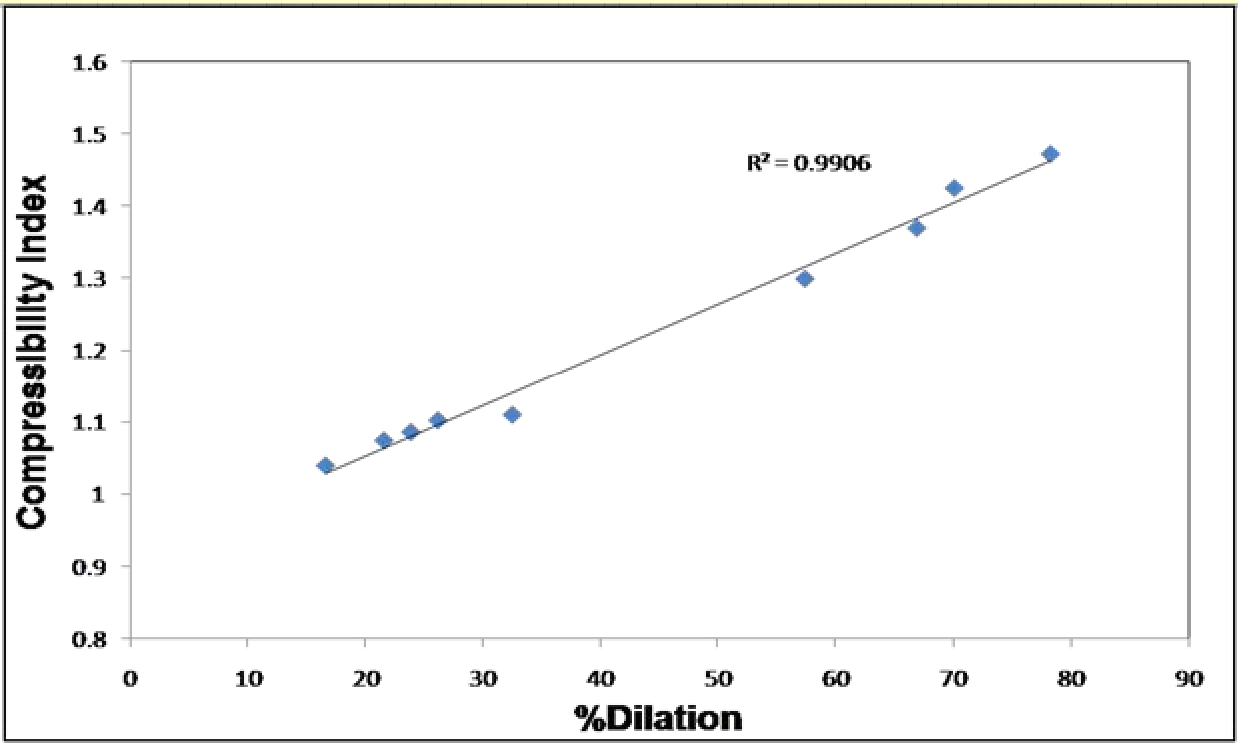
Compressibility



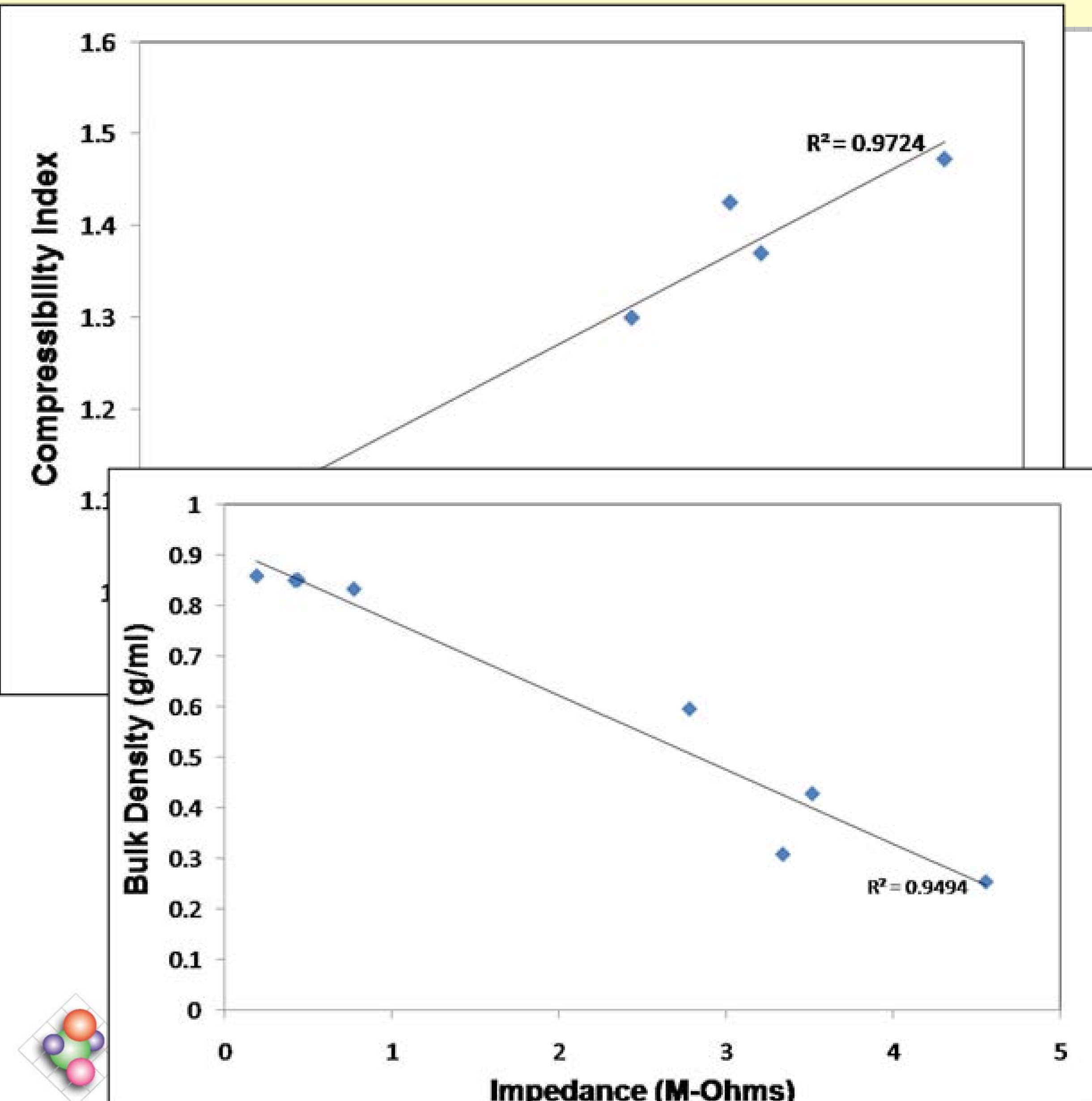
- *Dependence of Compressibility index on composition is very pronounced*

Compressibility vs. Dilation



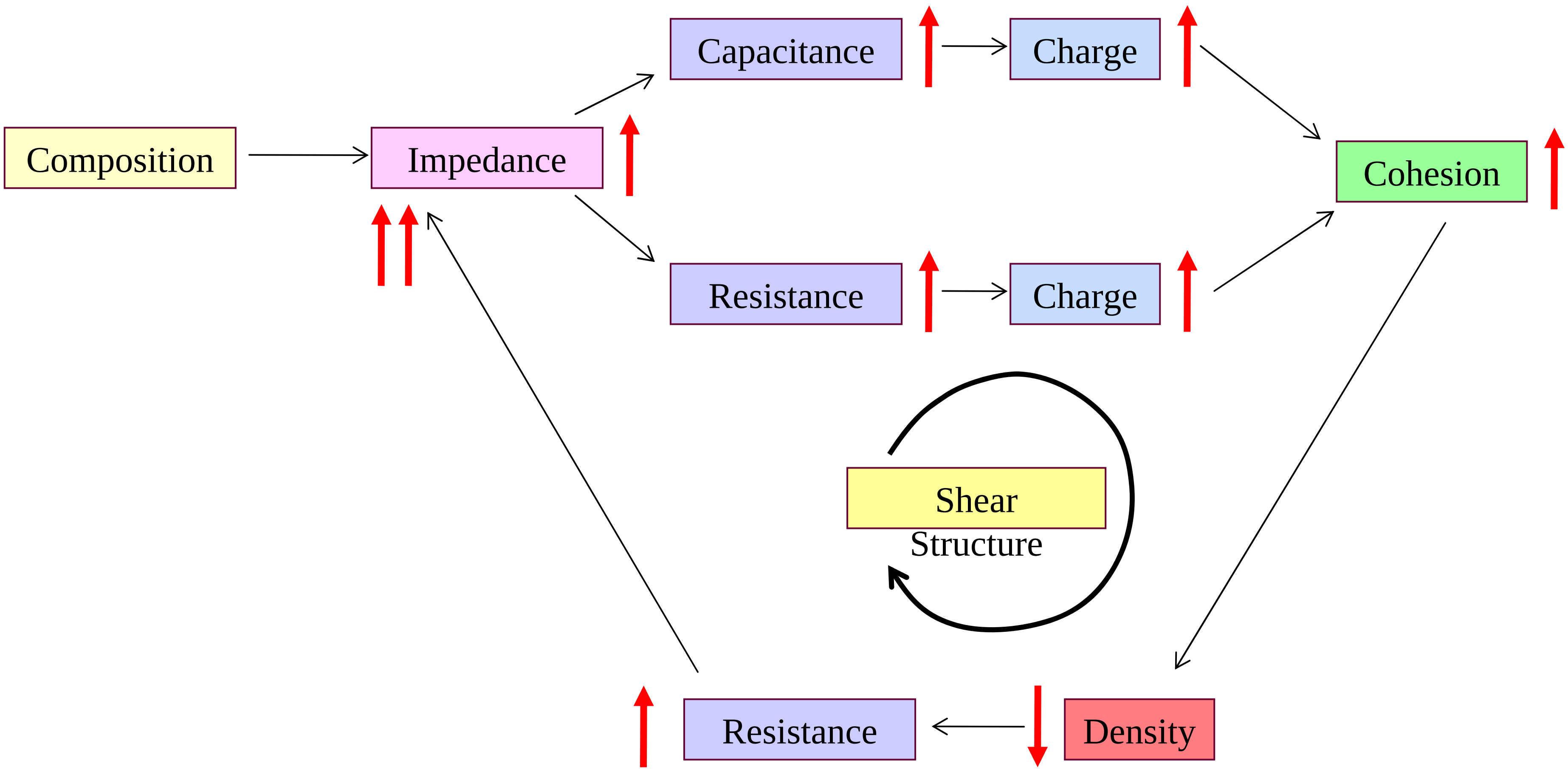
- *Flow properties of worst/intermediate flow blends are greatly influenced by the presence of highly compressible component*
- *High correlation between compressibility and dilation is present*

Impedance: Dependence on Density



- *Impedance values correlated well with compressibility index*
  - *Recall from previous (Reason #2), that lower density corresponds to larger voids, which results in lower conductivity and greater resistance of the powder bed*
- *To confirm this, impedance was also compared to <u>bulk density</u> measurements and showed good correlation*

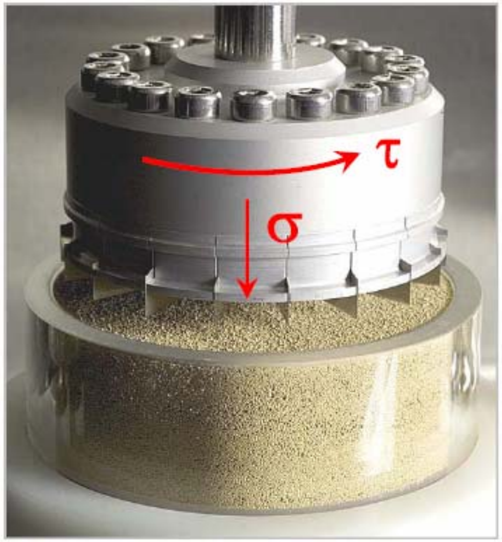
Relation Between Flow and Electrical Properties (Observations and Conclusions)



Methods: Rotating Shear Cell

- Application:
  - *Originally developed to assist in design of hoppers and silos*
  - *Subsequently used to characterize powder behavior in consolidated state*

- Procedure:
  - *Sample is loaded into a holding cup and pre-conditioned by dynamic blade*
  - *Sample is pre-compacted with a consolidation force S (stress σ)*
  - *Sample is pre-sheared to achieve critical consolidation state (steady-state flow)*
  - *Normal stress is lowered, sample is sheared further to obtain the yield point (a point of failure)*



FT4 Shear Cell
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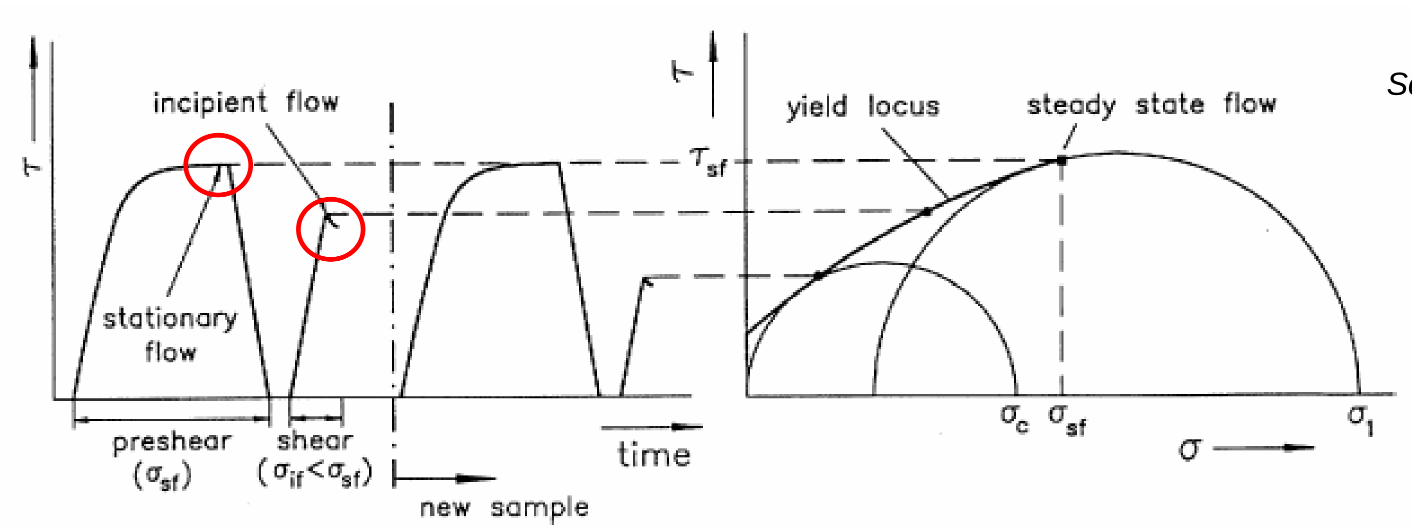
Methods: Rotating Shear Cell

- Yield Data:
  - *Procedure above is repeated several times to obtain <u>yield locus</u>*



Schwedes (1999)

  - *From yield locus, values of <u>major principal stress</u> (σ_1), <u>unconfined yield stress</u> (σ_c), and <u>cohesion</u> (the intercept) are obtained by Mohr stress circle analysis*

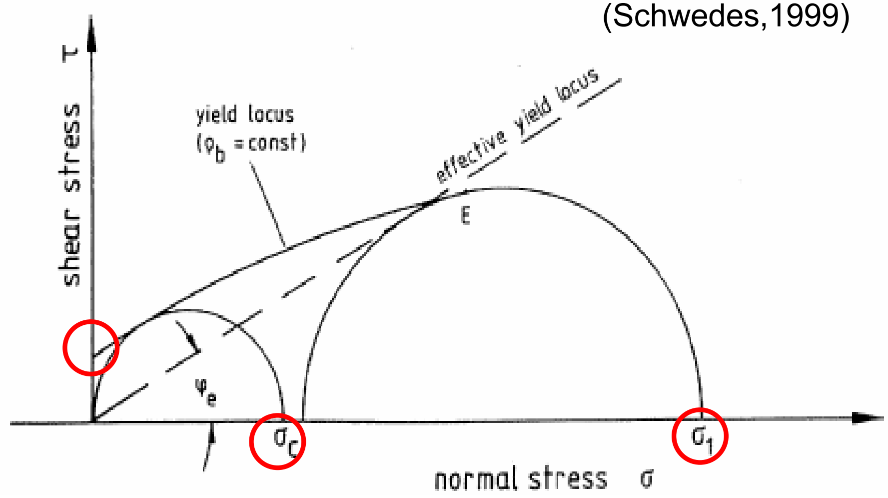
Methods: Rotational Shear Cell

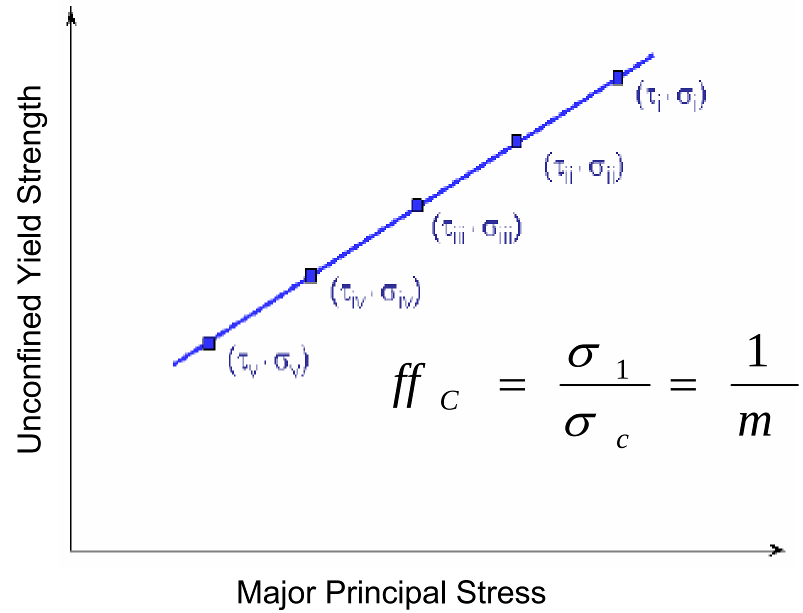
- Yield Data:
  - *σ_c was plotted against σ_1*
  - *Every blend was tested at 4 different consolidation stresses to obtain a flow function*

$$\sigma_c = f(\sigma_1)$$

  - *Flow factor (**ff_c**) was calculated for σ_N = 3 kPa (the smallest consolidation load tested)*
  - *Additionally, the value of cohesion parameter (value of shear stress at zero consolidation) was plotted against major principal stress (σ_1)*

(Schwedes,1999)





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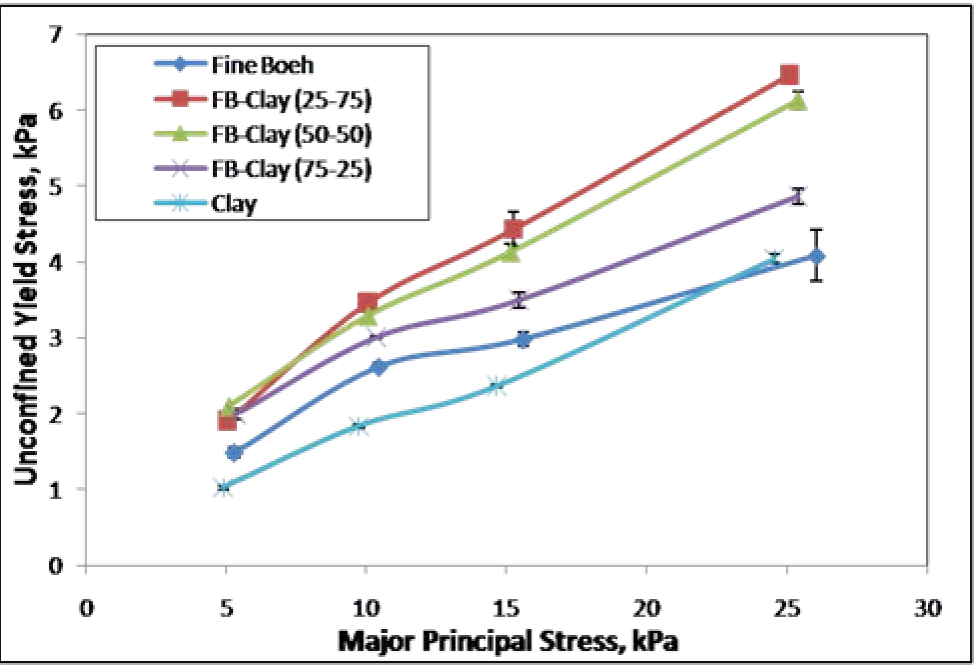
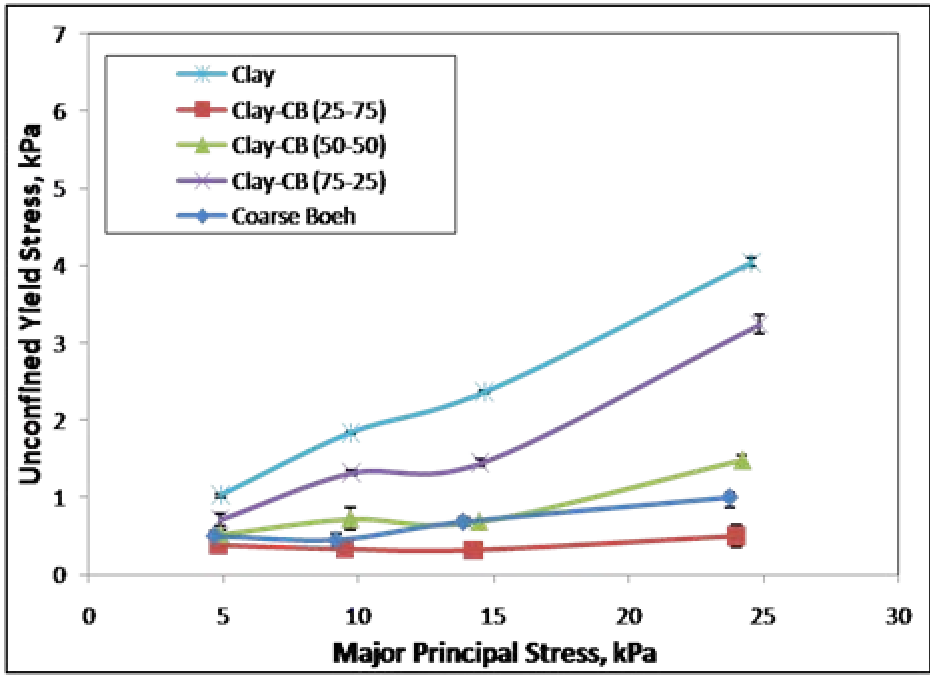
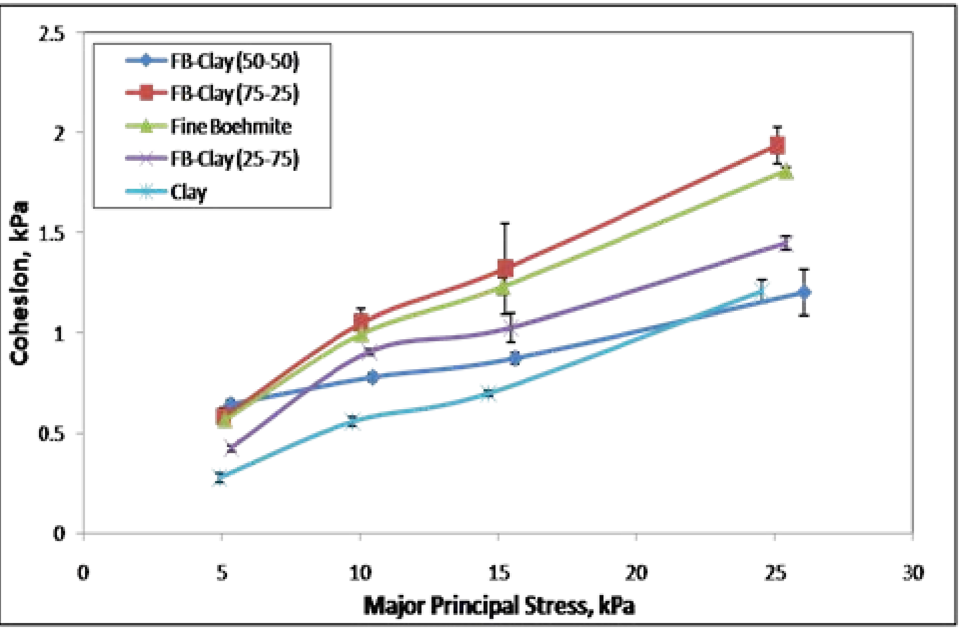
Shear Cell Data (Flow Functions)

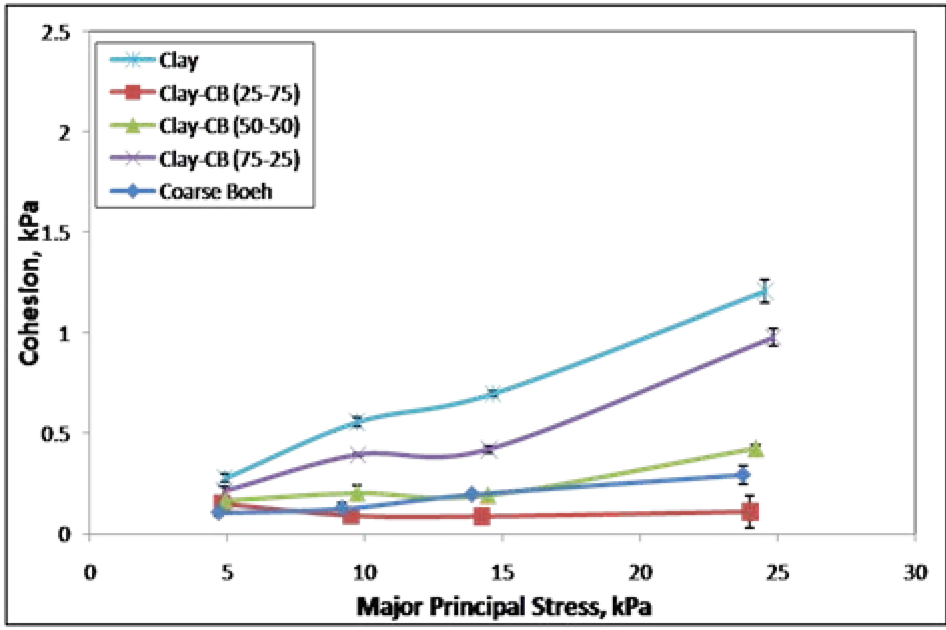




- ***<u>For FB/C mixtures</u>**: while the flow ranking was expected to be pure clay (best) → clay (75%) → clay (50%) → clay (25%) → FB (worst), all mixtures exhibited shear flow behavior that was worse than any of the pure components.*
- ***<u>For CB/C mixtures</u>**: shear behavior of mixtures was mostly as expected.*

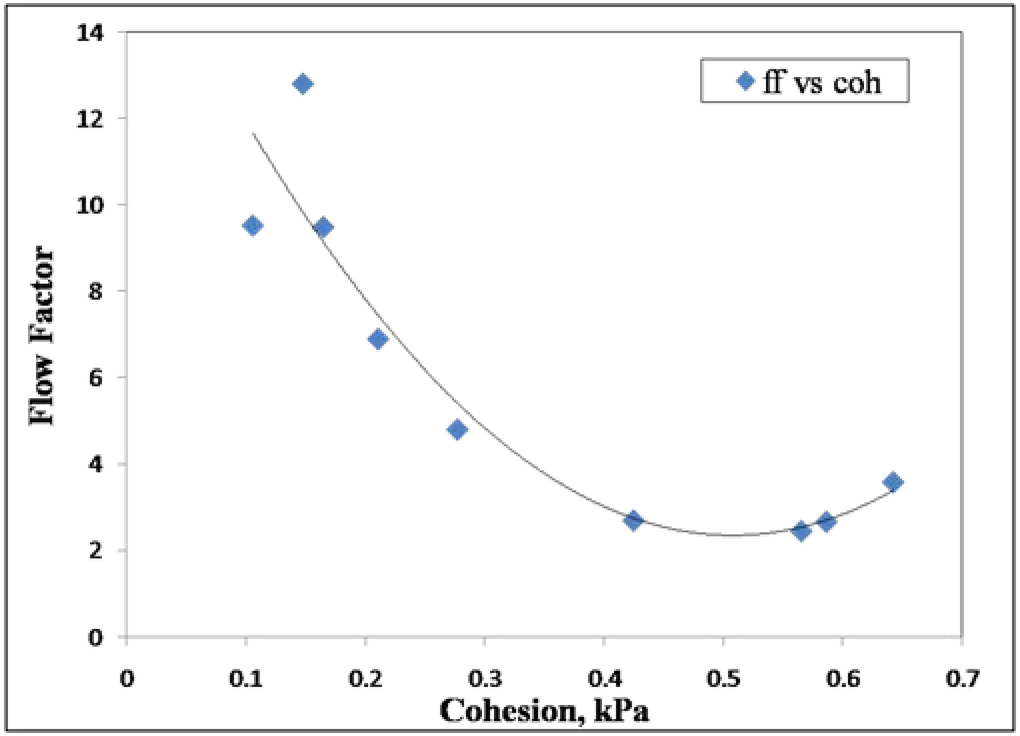
Shear Cell Data (Cohesion)





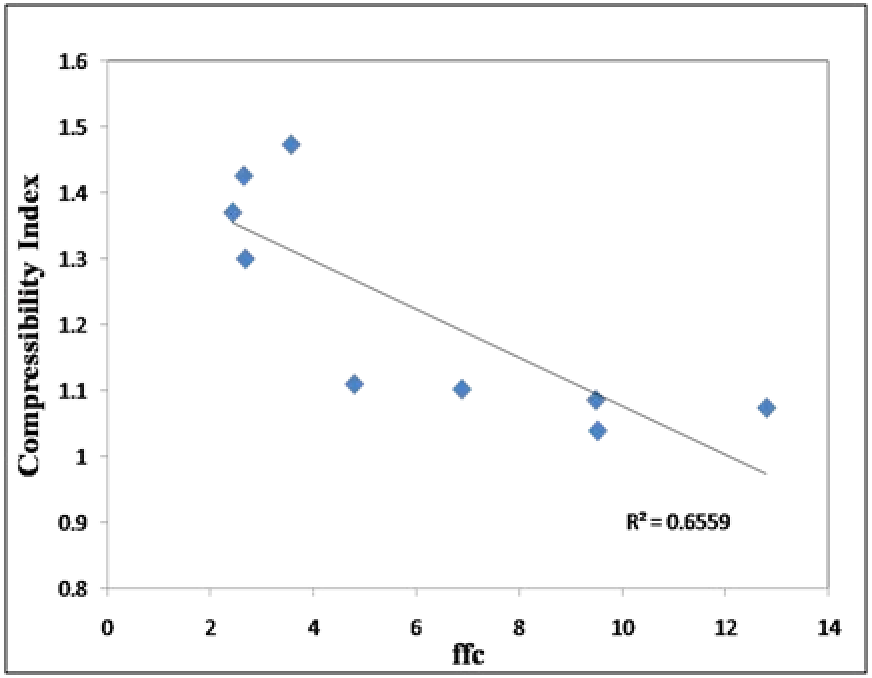
- ***For FB/C mixtures**: the flow ranking was expected to be pure clay (best) → clay (75%) → clay (50%) → clay (25%) → FB (worst), and most mixtures exhibited expected shear flow behavior.*
- ***For CB/C mixtures**: shear behavior of mixtures was mostly as expected.*

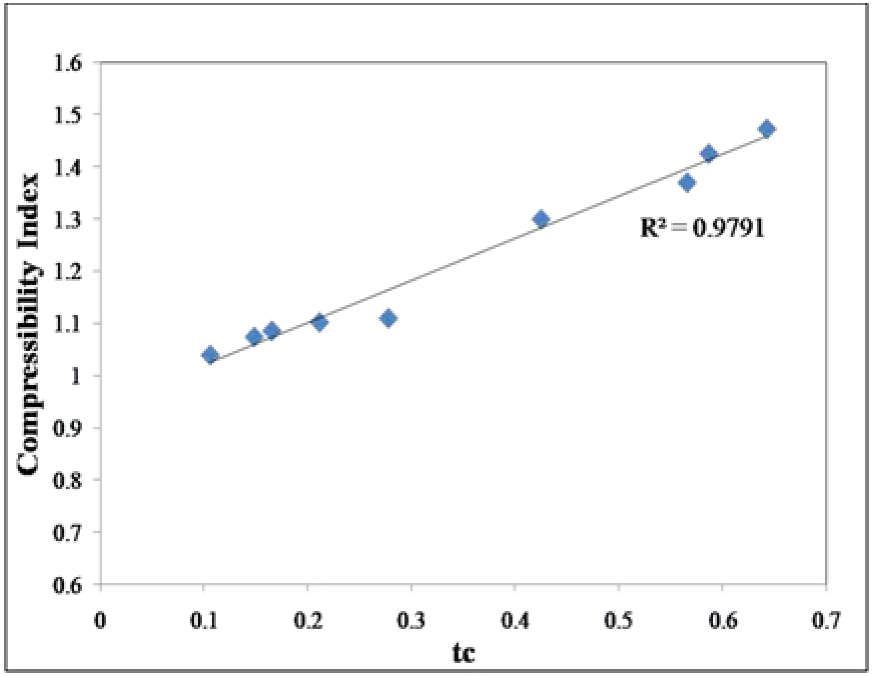
Shear Cell Results: Difference in Consolidation



- *Shear Cell Flow Factor (ff_c) and Cohesion parameter (τ_C) have a non-linear relationship;*
- *Cohesion parameter measures flowability under zero consolidation, while ff_c is much more consolidation-dependent*

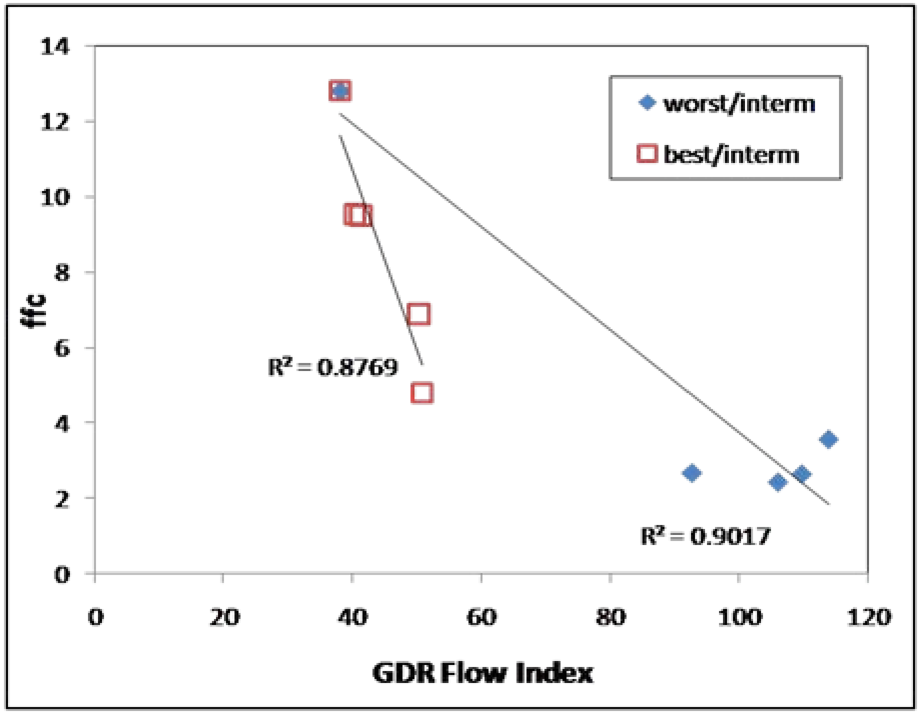
FT4 Techniques: Dependence on Consolidation

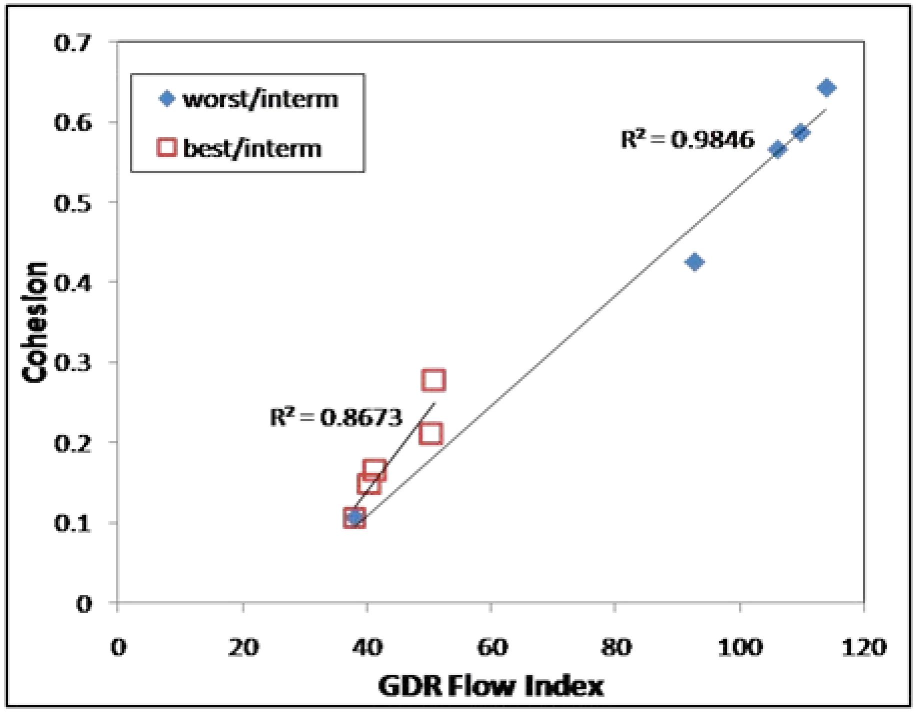




- *Shear Cell Flow Factor (ff_c) has a non-linear correlation to the Compressibility Index (I_C) due to the presence of cohesive components*
- *The correlation between I_C and cohesion is much more pronounced*

GDR vs. Shear Cell Correlations





- *Relationship between cohesion parameter (τc) and GDR Flow Index is quite more linear than for shear cell flow index (ff_c)*
- *Techniques measure flow characteristics at similar degrees of consolidation*

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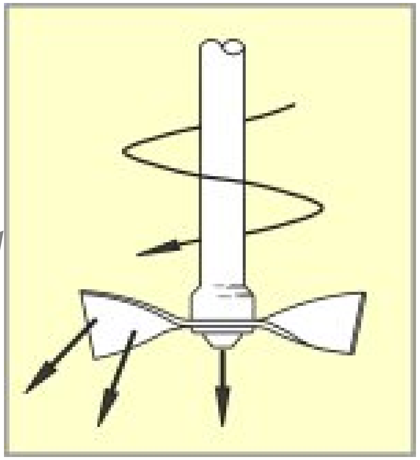
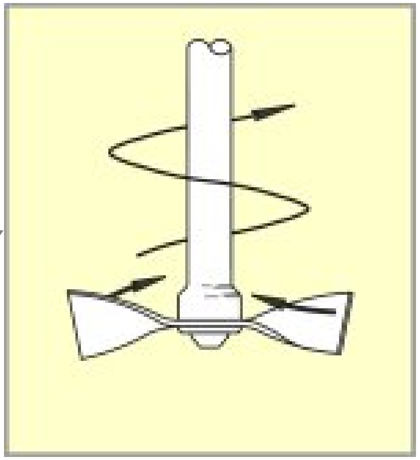
Conclusions (???)

- Higher impedance blends produce more cohesive beds that flow in larger "chunks" (i.e. produce higher flow indices) and dilate more.
- For materials with larger capacitance, capacitive charge storage increases the strength of inter-particle forces, leading to increases in cohesion.
- For more cohesive materials, decrease in density (increases in dilation) decreases electric conductivity.
- The powders with the best flow characteristics exhibited the lowest electrostatics (higher rates of discharge of electric charge are associated with improved flow).
- Higher dilation was mitigated by the presence of MgSt, which reduced the bed conductivity and dilation, and correspondingly improved the powder flow.
- Observed flow and electric properties are a complex function of blend composition.

Methods: Variable Flow Rate

- *Blade moves through the powder bed at decreasing flow rates (blade speed through the vessel)*
- *Several parameters are derived from the measurements:*
  - ***BFE (Basic Flowability Energy)*** *– energy measured with the blade passing downward through the bed (applying some compression)*
  - ***FRI (Flow Rate Index)*** *– ratio of flow energy of test 1 to flow energy of test 4 (sensitivity of powder to 10-fold decrease in flow rate)*
  - ***SE (Specific Energy)*** *– measures unconfined flow energy (energy on the blade moving upward)*





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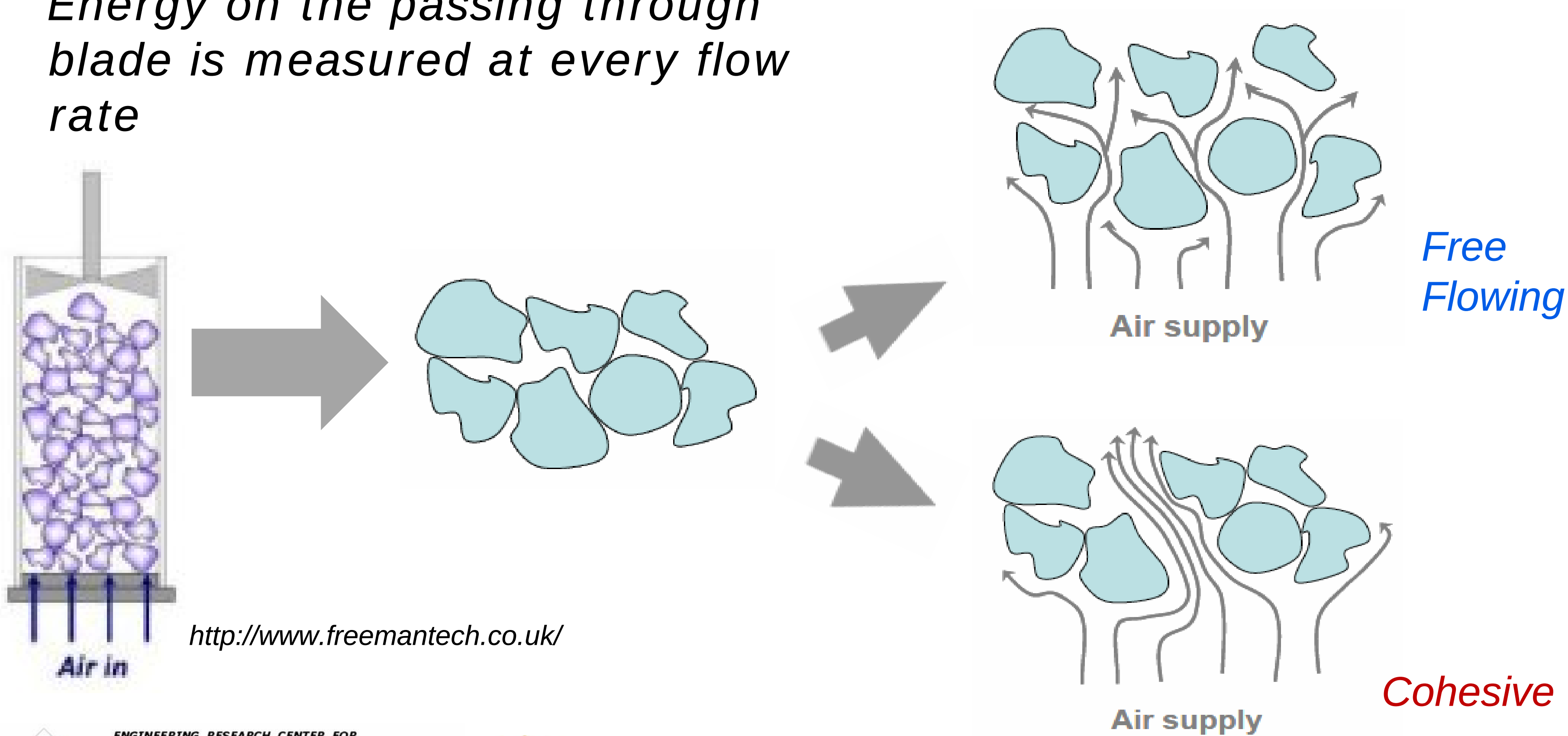
Methods: Aeration

- *Air is introduced through the bottom plate of the vessel at incremented intervals;*
- *Energy on the passing through blade is measured at every flow rate*
- *Measures how flow energy reduces with aeration*



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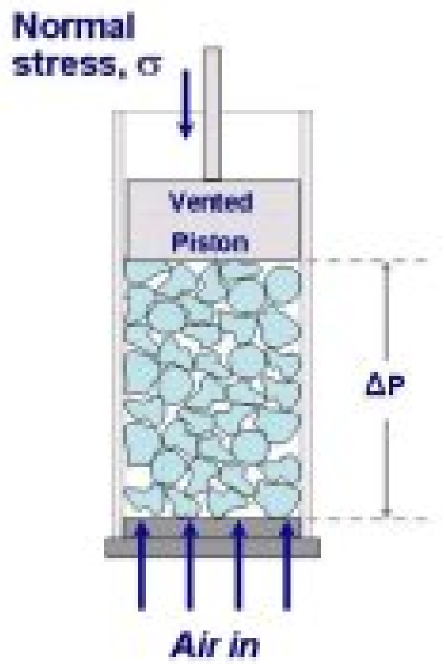
Methods: Permeability



- *Permeability measures how easily air passes through a powder at increasing bulk stress*
- *During the test the air is passed through the powder column base as the normal stress is applied in increments*
- *Pressure drop across the powder bed is measured, permeability constant (k) can be calculated*

$$Q = \frac{kA}{\mu}\frac{P_a - P_b}{L}$$

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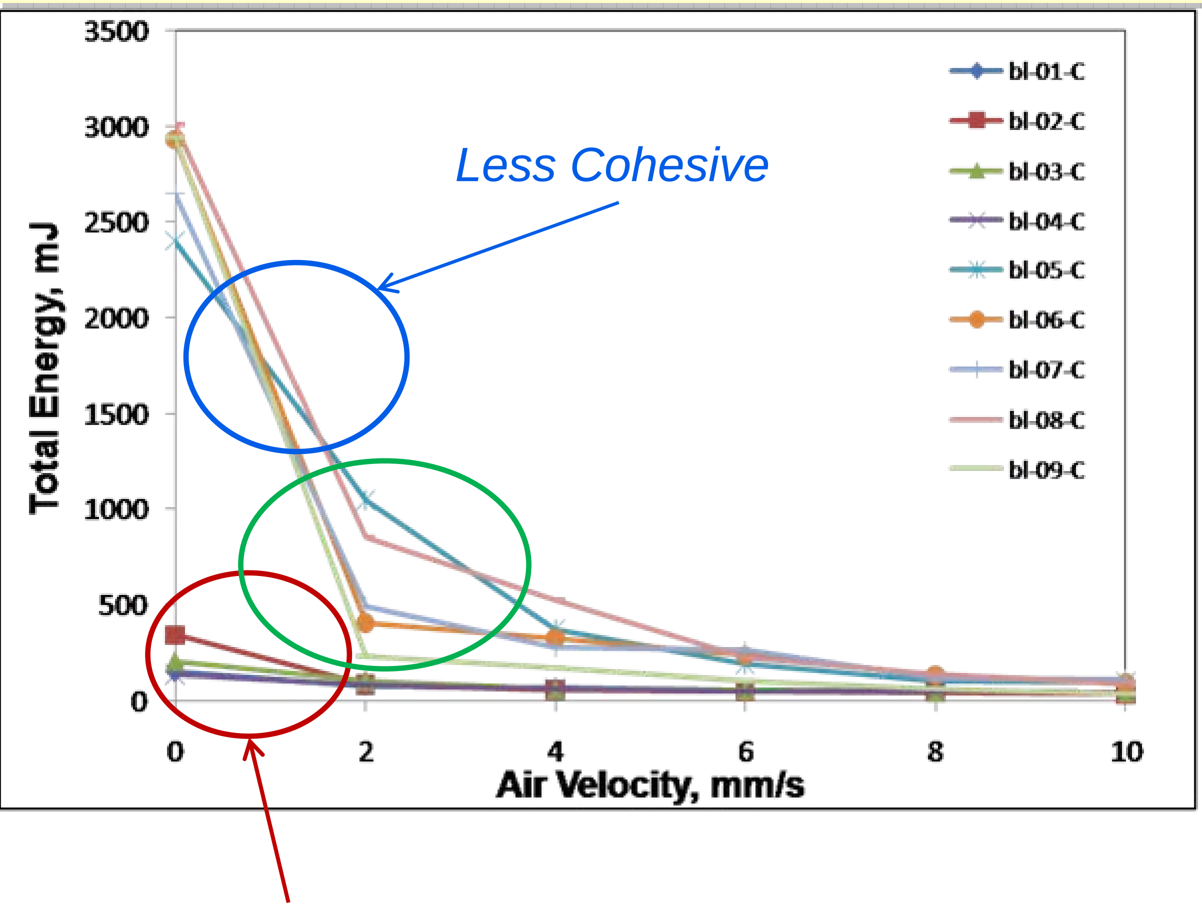
Where,
Q = air volume flow rate (cm^3/s)
k = permeability (cm^2)
A = cross-sectional area of bed (cm^2)
µ = air viscosity (Pa.s)
Pa-Pb = pressure drop across the bed (Pa)
L = length of powder bed (cm)

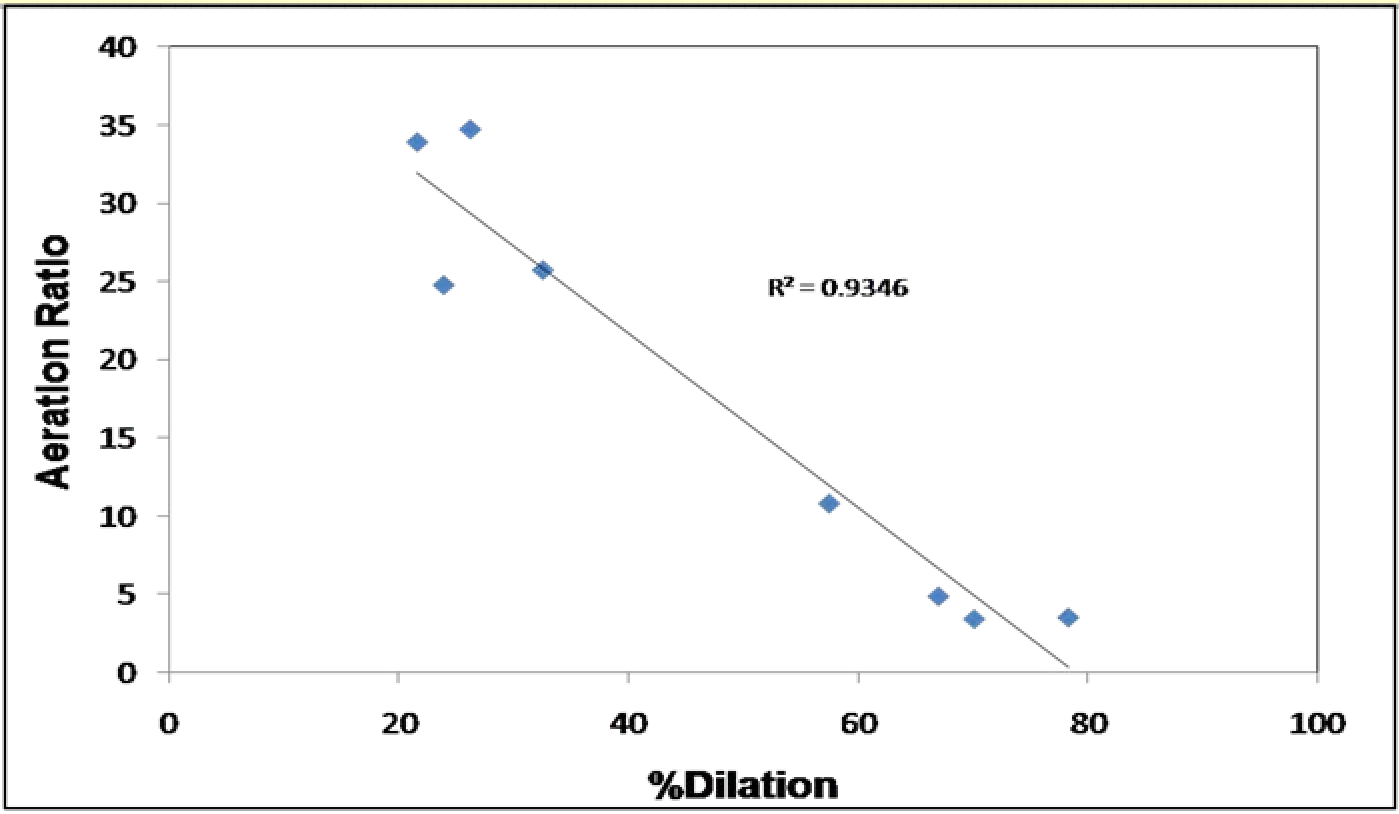
Aeration Results



More Cohesive

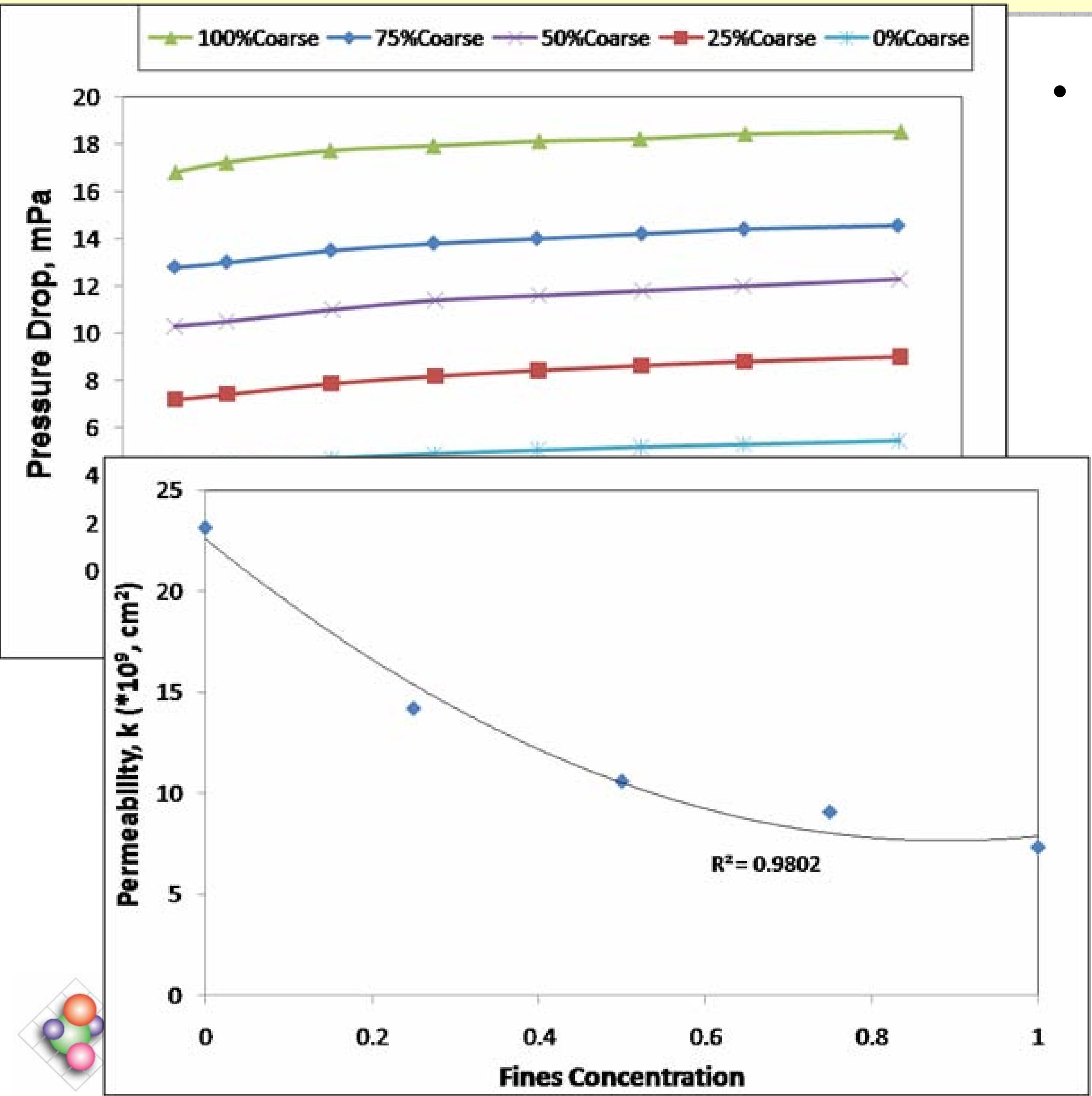
- *Blends containing <u>fine PSD ingredient</u> were hardly affected by aeration, with cohesive forces resisting fluidization*
- *Blends containing <u>coarse component</u> became fluidized quickly due to <u>weaker cohesive forces</u> between the particles*
- *The order, in which blends became fluidizes, follows the predicted order of blends cohesion (clay (worst) → 25% Coarse → 50% Coarse → 75% Coarse → 100% Coarse (best))*

Aeration and Dilation



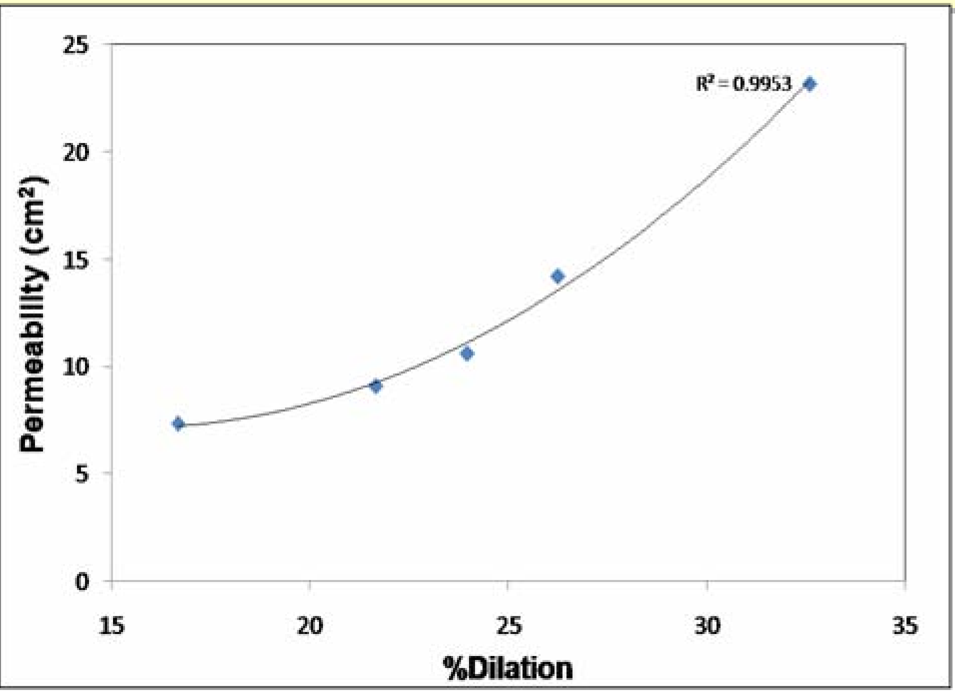
- *Lower aerated ratios tend to correspond to higher dilation volumes*
  - *Larger cohesive forces between the particles contribute to the formation of larger voids upon dilation; as well as, constrict the ability of air to pass through the powder bed and fluidize the powder*
  - *As a result, both these indices measure the same phenomenon*

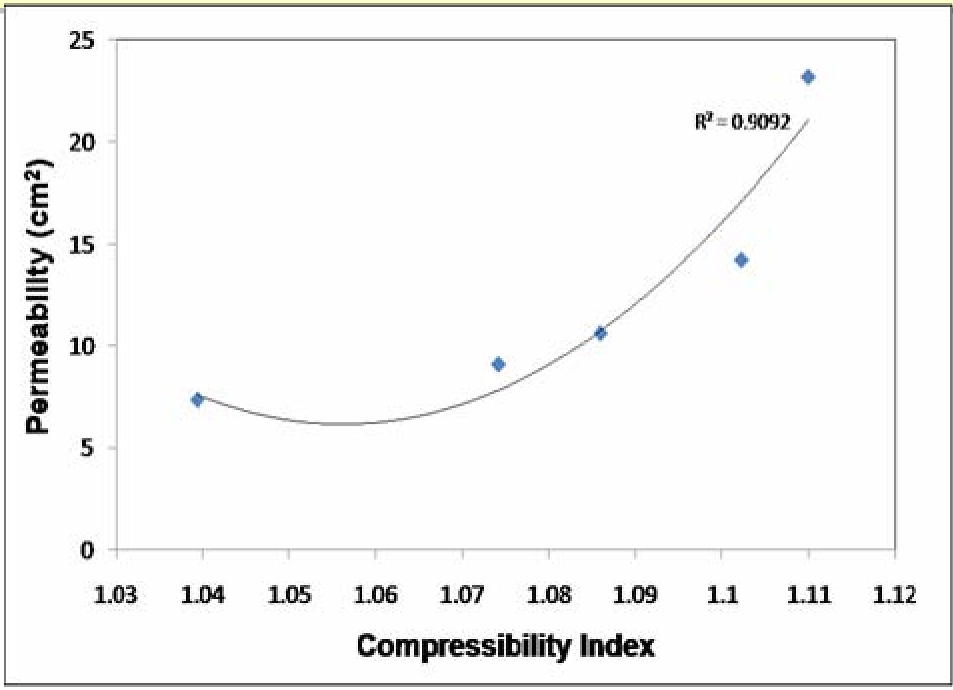
Permeability



- *Pressure drop (and permeability) showed to be a function of fines (clay) concentration*
    - *Trend of <u>decrease in permeability</u> with fines concentration is present, however, this dependence is non-linear*

Permeability: Consolidation





- *Previous finding also explain the non-linear trend between permeability and dilation (upper graph) and permeability and compressibility index, suggesting a combination of other factors may influence the permeability results*