Application of Interparticle Spacing Model to Maximize Filler Content in Concrete

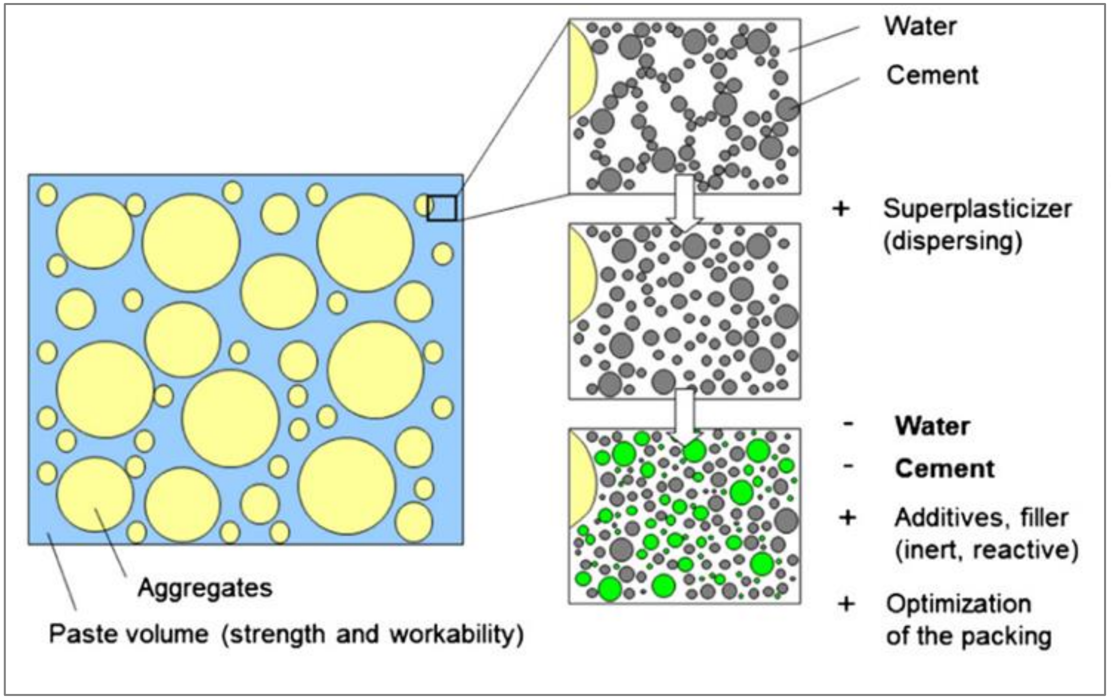
Lead:	**ORNL (Denise A. Silva)**
NL partner:	**PNNL**
Academic partners:	**University of São Paulo, Brazil** **University of Tennessee, Knoxville**
Industrial partners:	**Precast Concrete Institute** **GCP Applied Technologies** **Four precast concrete producers**
Sponsor:	**U.S. Department of Energy (BTO, IEDO)**

October 31, 2023

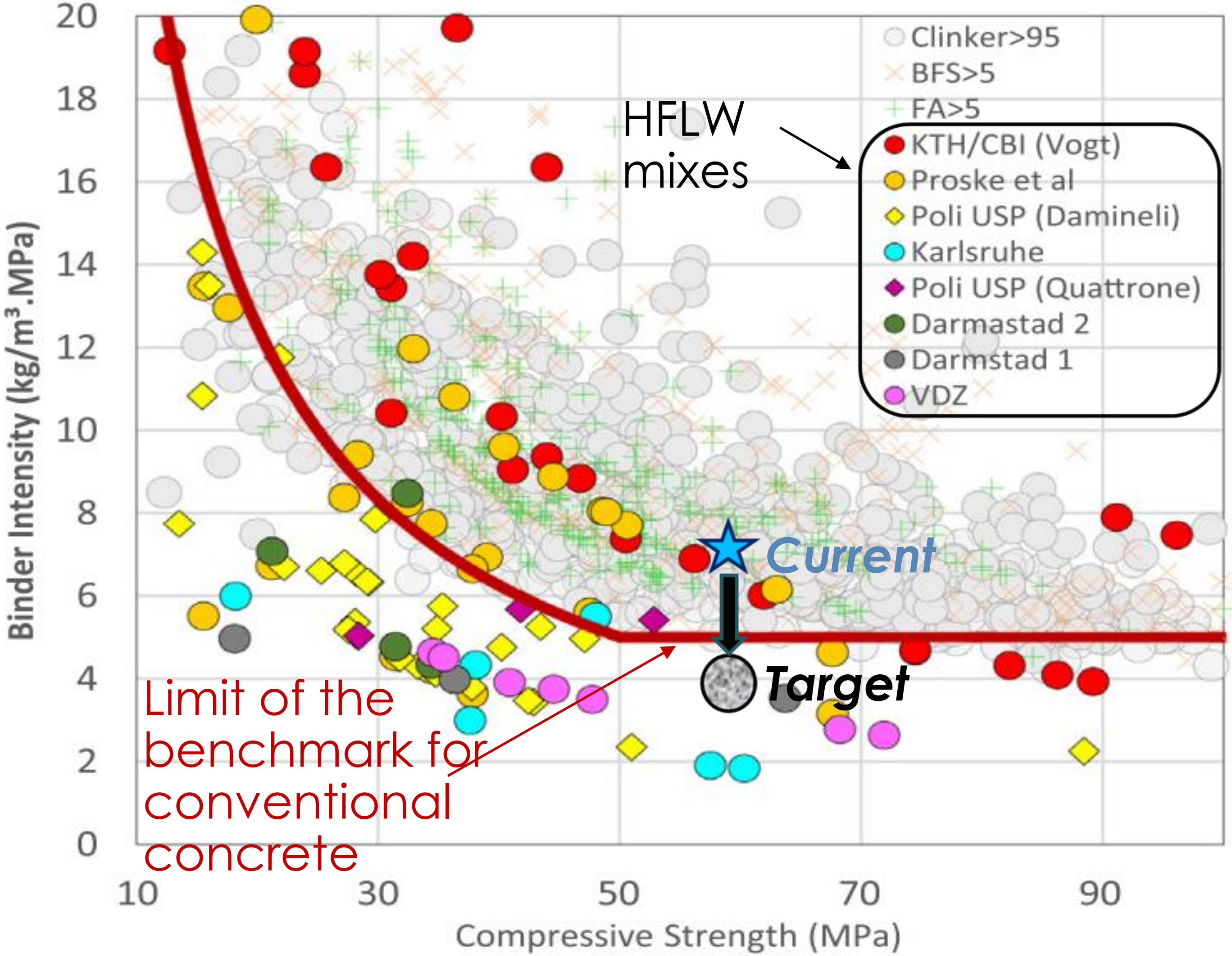
Introduction

- Cement use efficiency improvement & cement replacement by SCMs can enable the cement/concrete industry to significantly reduce carbon emissions in the near term.
- Cement use efficiency can be improved by minimizing the porosity of the granular skeleton of the concrete system, allowing up to 70% and 50% reduction in cement and water consumptions, respectively, for similar performance and cost.
- Filler particles partially replace cement grains and fill voids between cement particles, using particle packing models as a tool: **High Filler, Low Water (HFLW) concrete** (John et al., 2018).
- Higher packing density impacts concrete flow: use of rheometry and rheological models to achieve adequate workability.
- Here, the initial steps taken to implement the HFLW technology in a U.S. precast/prestress concrete producer are described.

The HFLW Concrete Technology



Proske et al, 2013



HFLW mixes

Current

Target

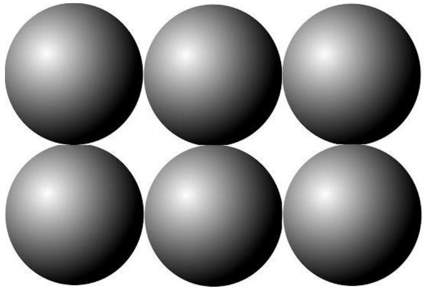
Limit of the benchmark for conventional concrete

John et al., CCR 2018

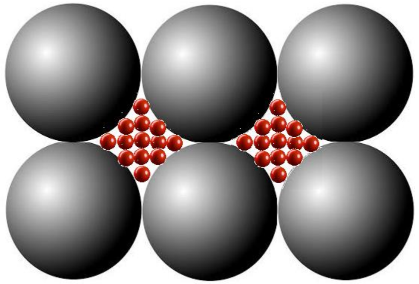
Particle Packing and Mobility Models

- **Westman & Hugill's algorithm (1930):** apparent volume V_a of the granular system with the highest volume of pores (worse situation).

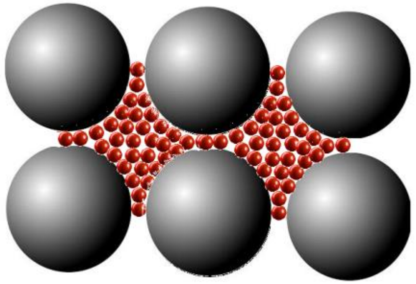
$$\begin{aligned} V_{a1} &= a_1 x_1 \\ V_{a2} &= x_1 + a_2 x_2 \\ V_{a3} &= x_1 + x_2 + a_3 x_3 \\ &\vdots \\ V_{ai} &= \sum_{j=1}^{i-1} x_j + a_i x_i \\ &\vdots \\ V_{an} &= \sum_{j=1}^{n-1} x_j + a_n x_n \end{aligned}$$



Compositions near 100% coarse: Va of mixture determined by the coarse particles.



Mixture pore volume Pv decreases when fines fit into pores between coarse particles. Minimum mixture Pv: coarse Pv = fine bulk volume (Bv).



If Va fines > pore volume of coarse particles: coarse particles dispersed within bulk volume of fines.

Va = Va of fines + true coarse volume

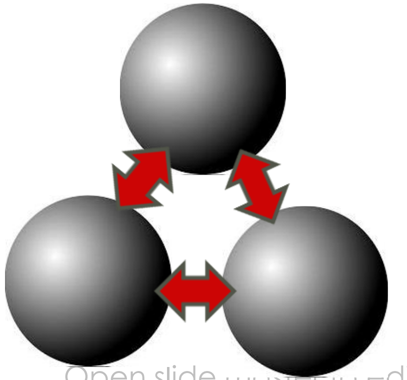
- **Funk and Dinger (1994):** interparticle spacing model for fine and coarse concrete fractions (IPS, MPT).

$$IPS = \frac{2}{VSA} x \left[\frac{1}{\phi s} - \left(\frac{1}{(1-Pof)}\right)\right]$$

$$Pof = 100\% \left[1 - \frac{1}{Va}\right] * 0.4$$

$$MPT = \frac{2}{VSA_C} \times \left[\frac{1}{V_{SC}} - \left(\frac{1}{1-P_{of_C}}\right)\right]$$

- *P_{of-}- porosity in max packing condition*
- *P_{ofc} - paste volume*



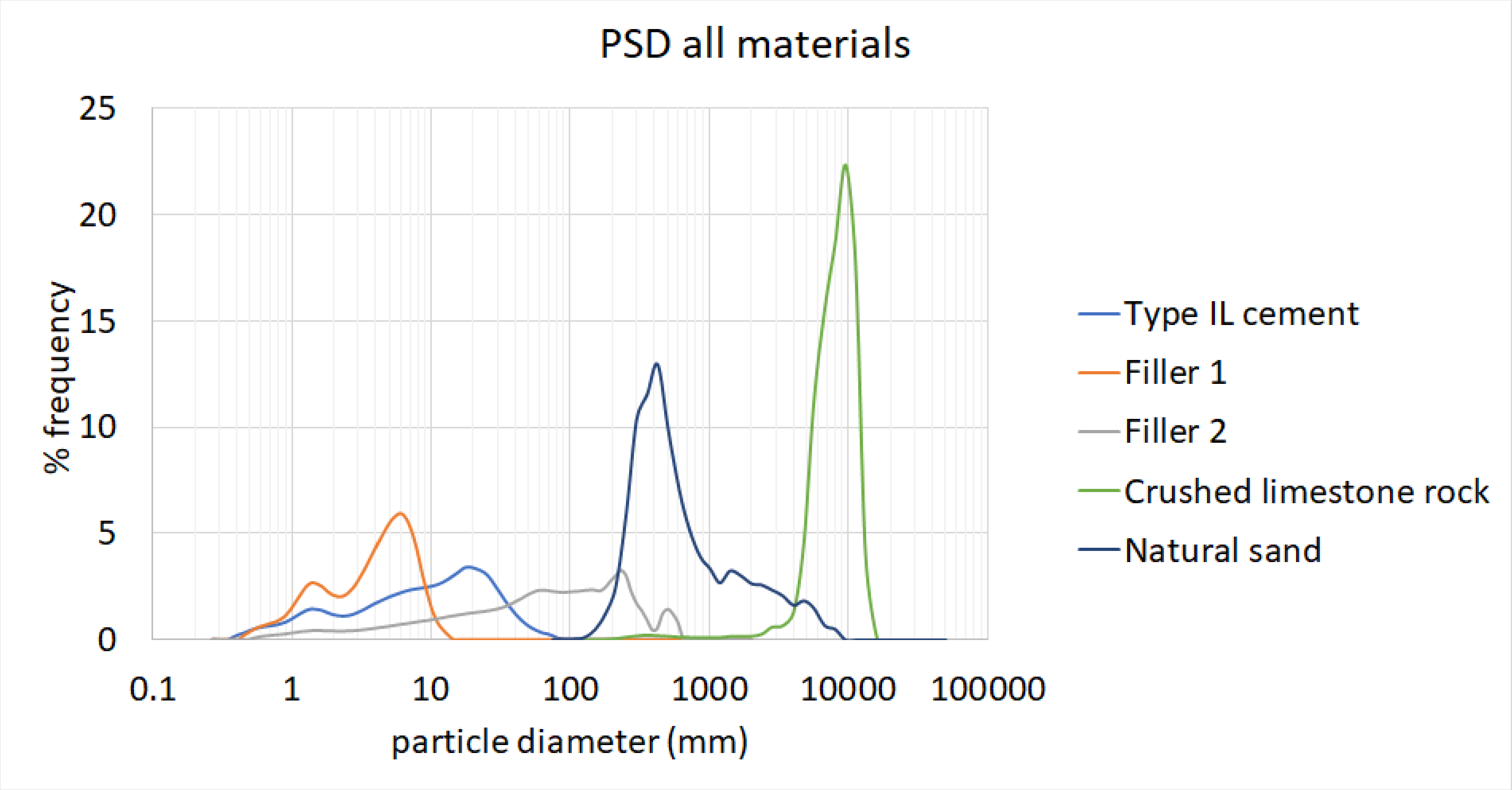
Materials

- ASTM Type IL Portland Cement (Portland Limestone Cement or PLC)
- Filler 1: calcitic, limestone, finer
- Filler 2: dolomitic limestone, coarser
- Natural quartz sand
- Crushed limestone coarse aggregate
- Polycarboxylate ether (PCE)-based dispersant

Parameter	*PLC*	*Filler 1*	*Filler 2*
SiO_2 (%)	18.44	1.31	1.3
Al_2O_3 (%)	4.07	0.22	0.24
Fe_2O_3 (%)	3.05	0.09	0.15
CaO (%)	62.16	53.74	30.4
MgO (%)	2.15	0.63	20.13
SO_3 (%)	3.18	0.02	0.23
Na_2O (%)	0.09	0.01	<0.01
K_2O (%)	0.52	0.02	0.04
L.O.I. (%)	5.62	43.40	46.87
$D(10)$ (μm)	1.225	1.097	4.9
$D(50)$ (μm)	9.22	3.960	65.72
$D(95)$ (μm)	37.68	8.510	395.5
Mean ϕ (μm)	13.93	4.130	111.5

Parameter	*PLC*	*Filler 1*	*Filler 2*
C_3S (alite) (%)	63.1	-	-
C_2S (belite) (%)	7.9	-	-
Cubic C_3A (%)	3.1	-	-
C_4AF (ferrite) (%)	9.3	-	-
Gypsum (%)	3.7	-	-
CaO (free lime) (%)	1.3	-	-
$Ca(OH)_2$ (portlandite) (%)	0.6	-	-
MgO (periclase) (%)	1.4	-	-
$CaCO_3$ (calcite) (%)	8.6	96.3	1.2
$MgCa(CO_3)$ (dolomite) (%)	0.5	0.5	98.4
SiO_2 (quartz) (%)	0.7	3.2	0.4
BET SSA (m^2/g)	1.37	2.00	0.71
True density (g/cm^3)	3.068	2.831	2.920

Techniques: Sieving, QXRD, XRF, laser diffraction PSD, N_2 adsorption (BET) for SSA, He-pycnometry

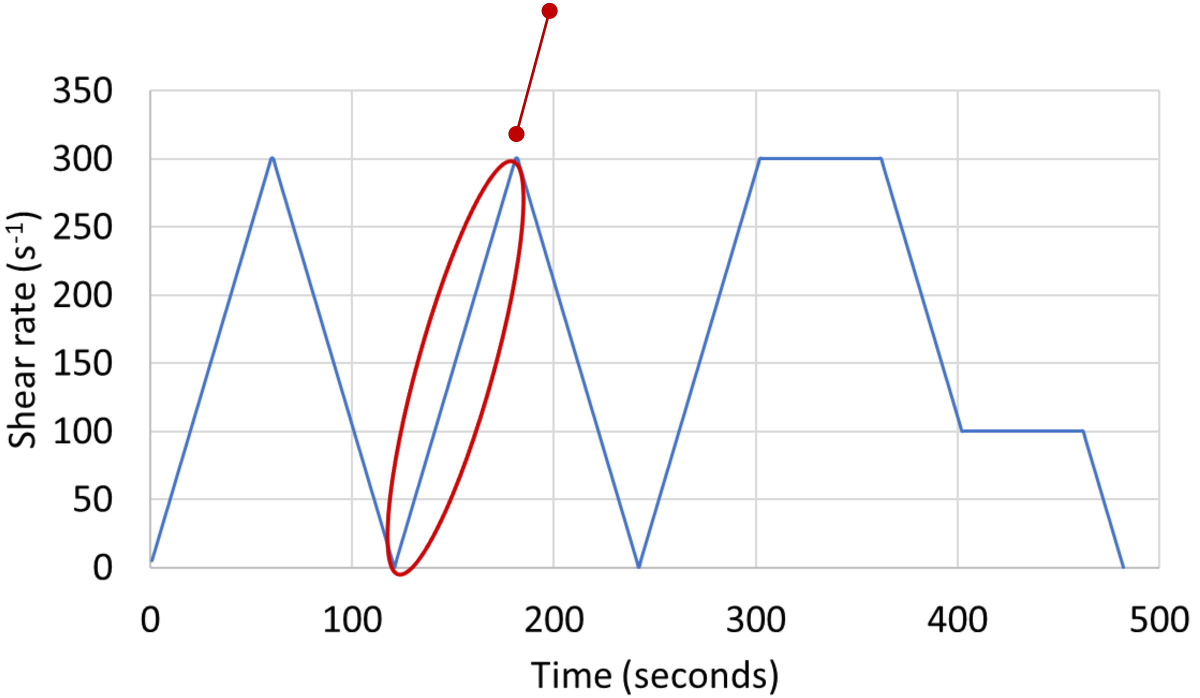

PSD all materials
% frequency
25
20
15
10
5
0
0.1
1
10
100
1000
10000
100000
particle diameter (mm)
Type IL cement
Filler 1
Filler 2
Crushed limestone rock
Natural sand

Experimental

DEFINITION OF CONCRETE FINE FRACTION

1. Determination of dispersant requirement for full dispersion of unitary pastes.
2. Determination of impact of filler replacement in composite pastes.
3. Techniques:
   - ❖ Bob & cup rheometry at 23°C (4,000-10,000 rpm mixing per ASTM C1738)
   - ❖ Isothermal calorimetry
4. Estimation of IPS of composite pastes using particle packing models.

Concentric cylinders rheometry

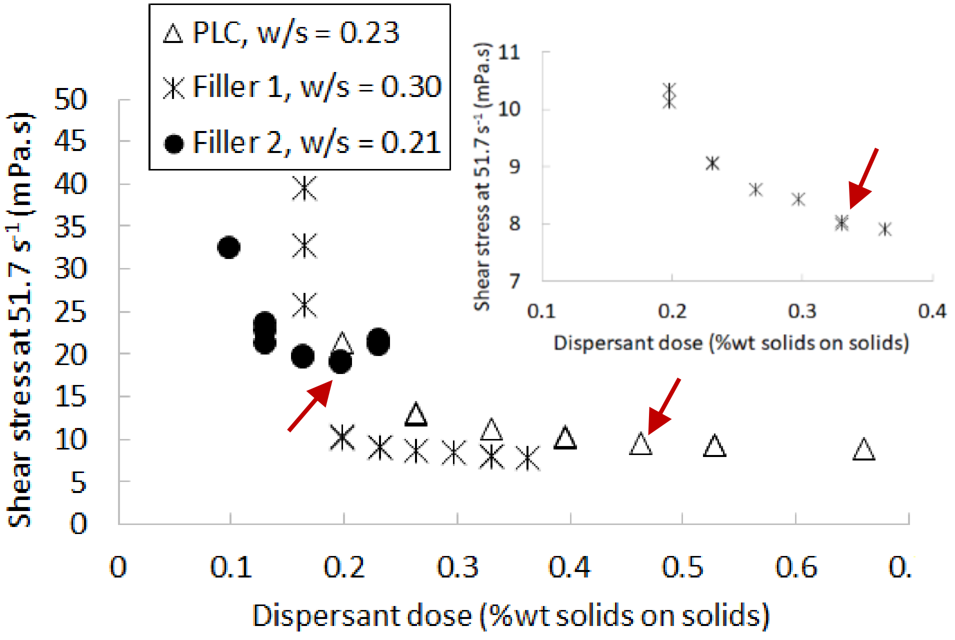

Apparent viscosity and rheology models applied to 3rd shear rate interval

DEFINITION OF CONCRETE COARSE FRACTION

1. Starting point is the reference concrete.
2. Estimation of MPT of concrete mixes for absolute volume and similar paste volume.
3. Concrete lab testing including rheometry.

Results - Fine Fraction

Rheometry of unitary pastes



(a)

- Calculated content of dispersant for maximum dispersion of binary and ternary pastes: 0.33% - 0.42% s/s (weighed average)

Results - Fine Fraction

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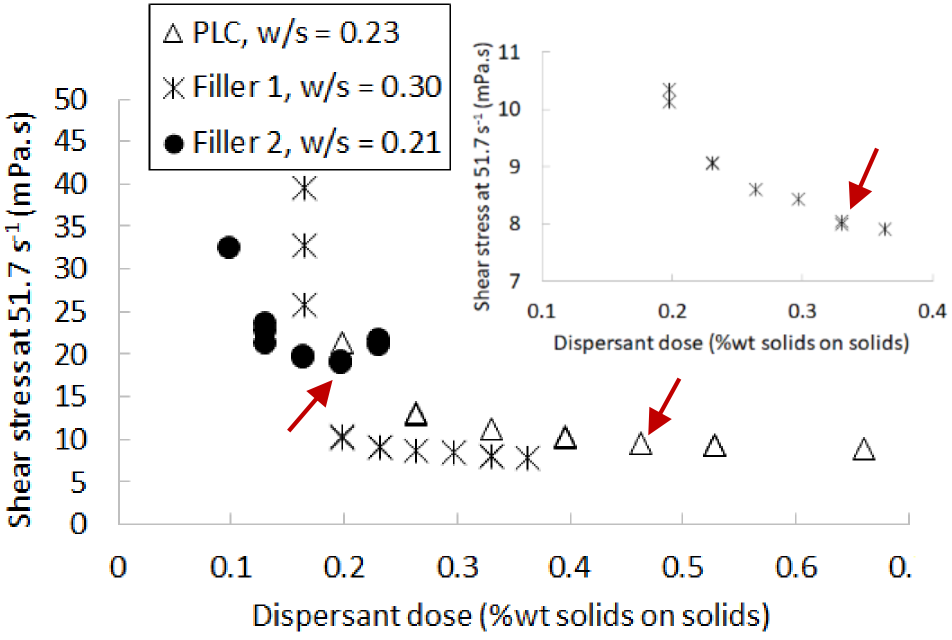
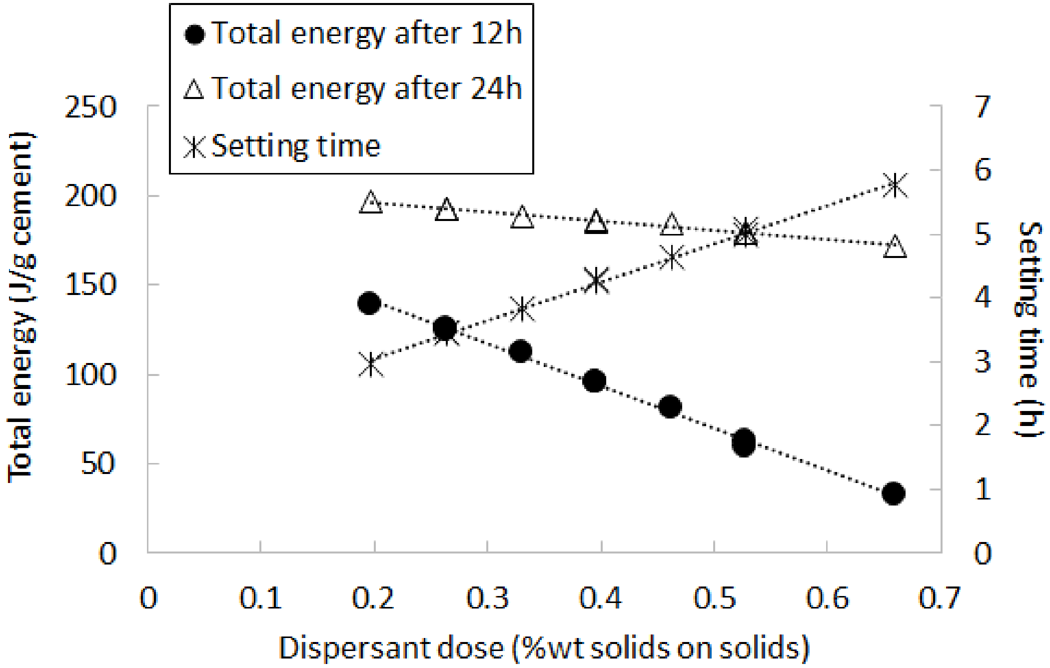


(a)

Calorimetry of PLC pastes, w/s = 0.23



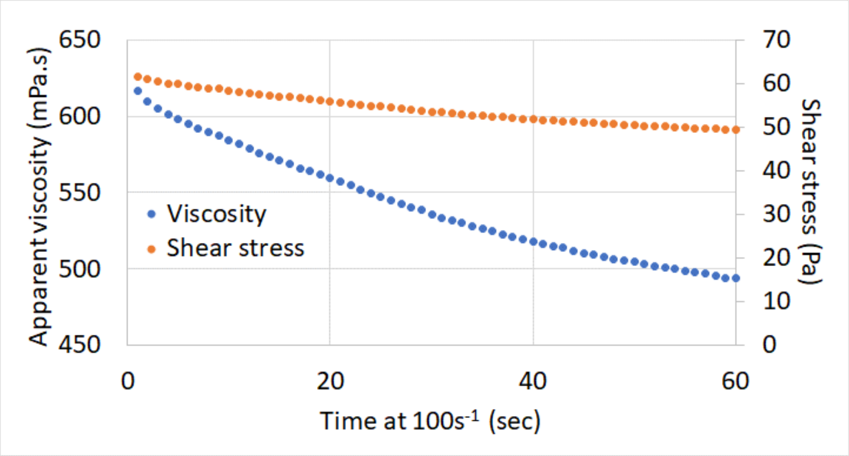
(b)

- Calculated content of dispersant for maximum dispersion of binary and ternary pastes: 0.33% - 0.42% s/s (weighed average)
- Choice of dispersant dose should consider performance (rheologic behavior, hydration kinetic parameters) and cost: 0.33% s/s

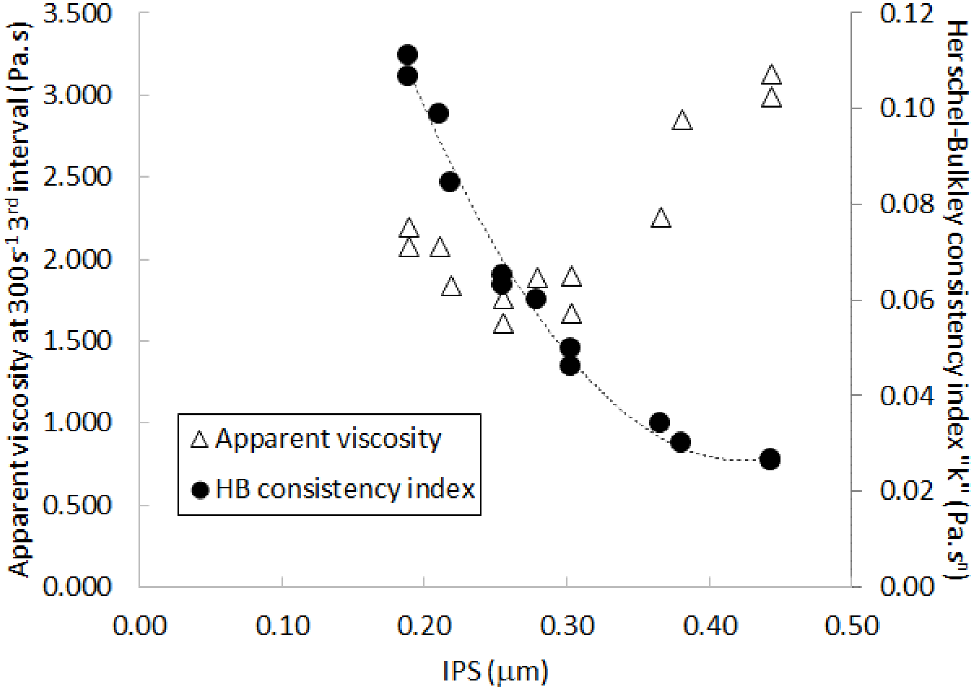
Source: Silva et al., 2023 (16th ICCC Proceedings)

Results - Fine Fraction

- Lack of correlation IPS x apparent viscosity because pastes are not in equilibrium under testing conditions.



- Correlation IPS x Herschel-Bulkley consistency index.



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Results - Fine Fraction

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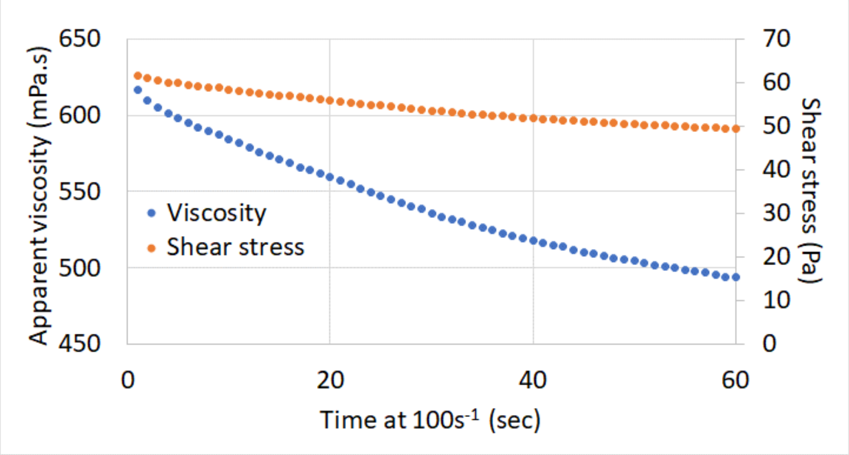
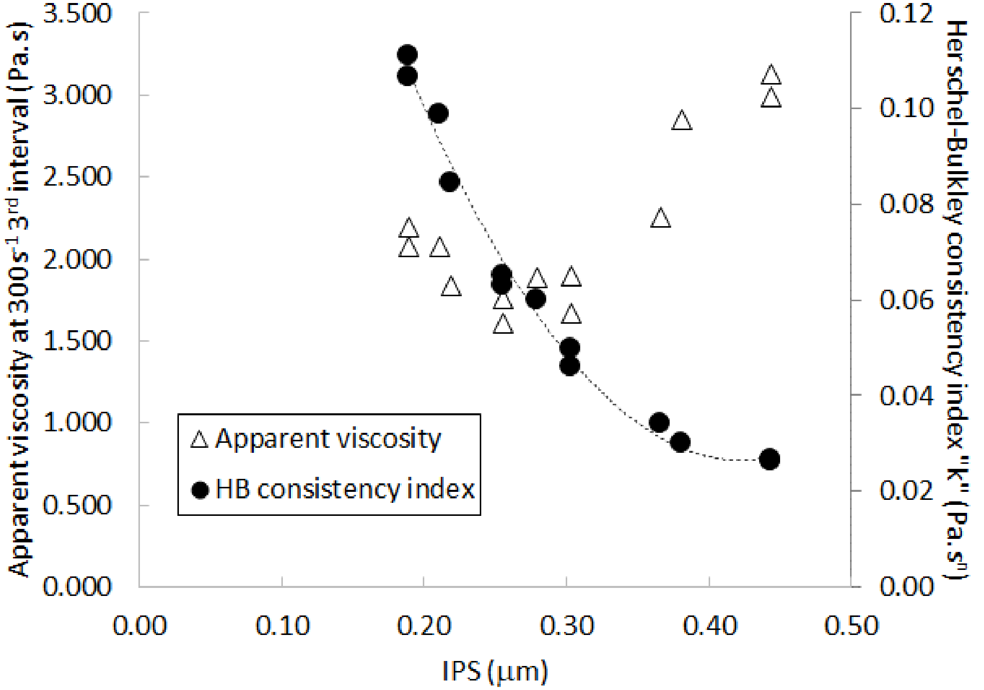
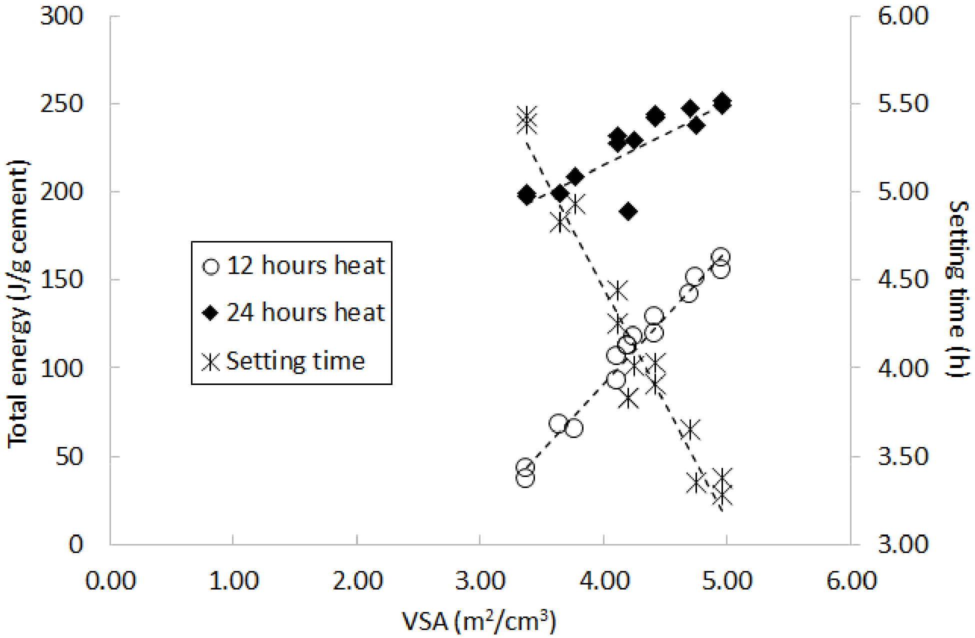


- Correlation IPS x Herschel-Bulkley consistency index.
- Good correlation between particle packing, rheological parameters and kinetic parameters





OAK RIDGE
National Laboratory

Results – Fine Fraction

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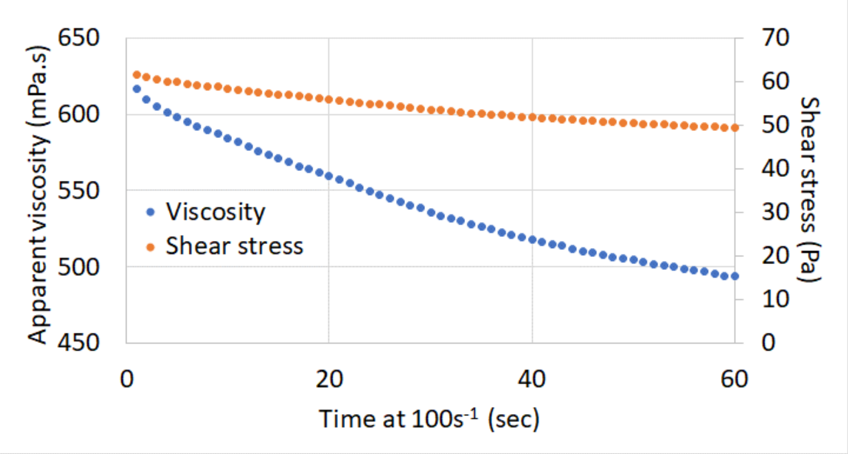
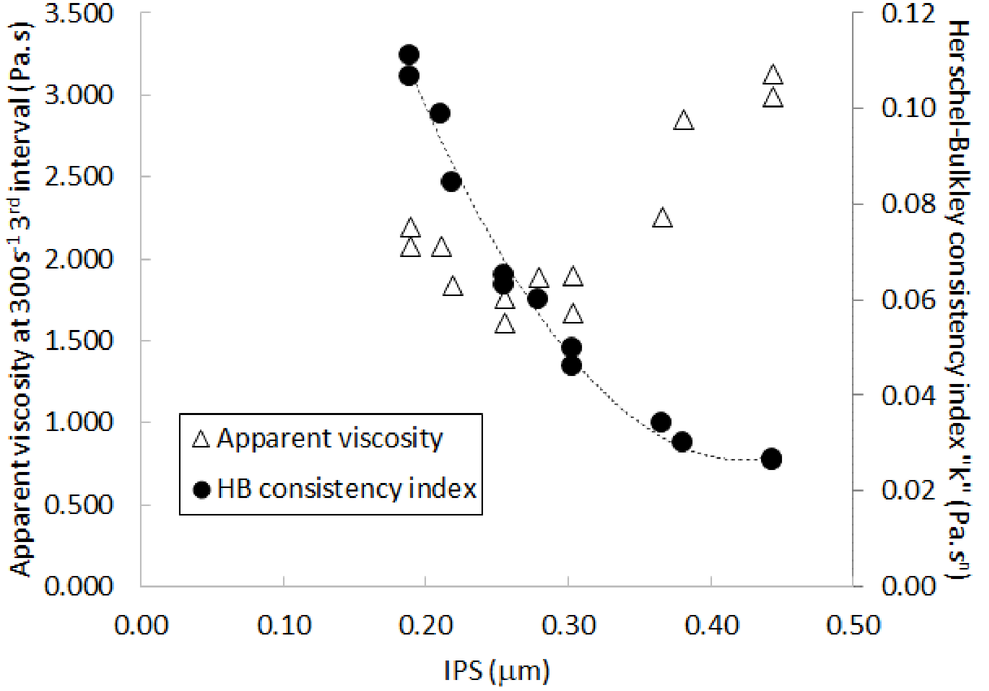


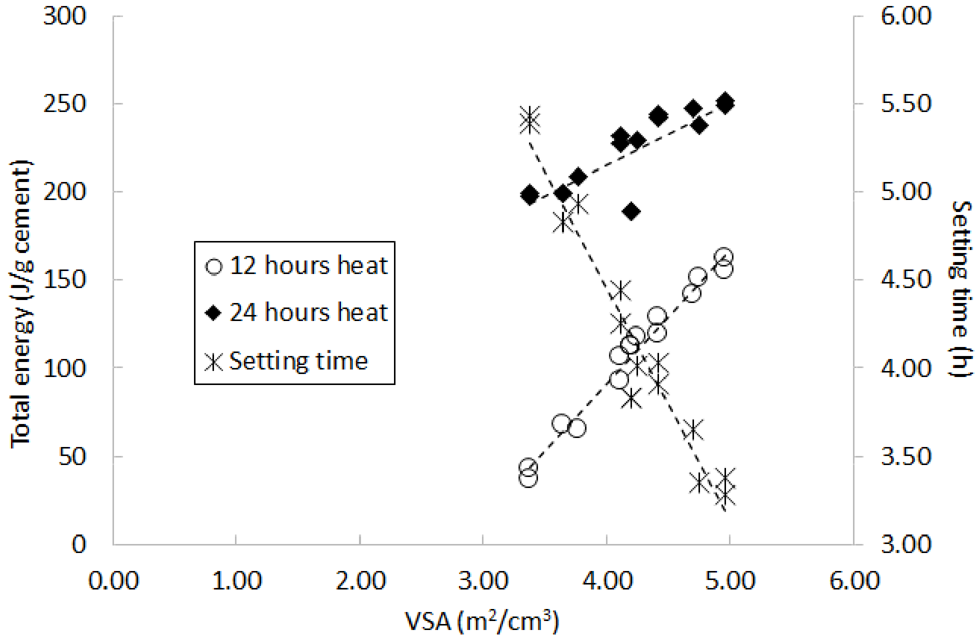
- Correlation IPS x Herschel-Bulkley consistency index.
- Good correlation between particle packing, rheological parameters and kinetic parameters

✓ IPS is a good indicator of HFLW pastes rheological behavior, with potential to design low carbon pastes.

✓ Strong correlation between hydration kinetics, SSA, IPS.

✓ A balance between rheology adequacy and hydration kinetics is critical.

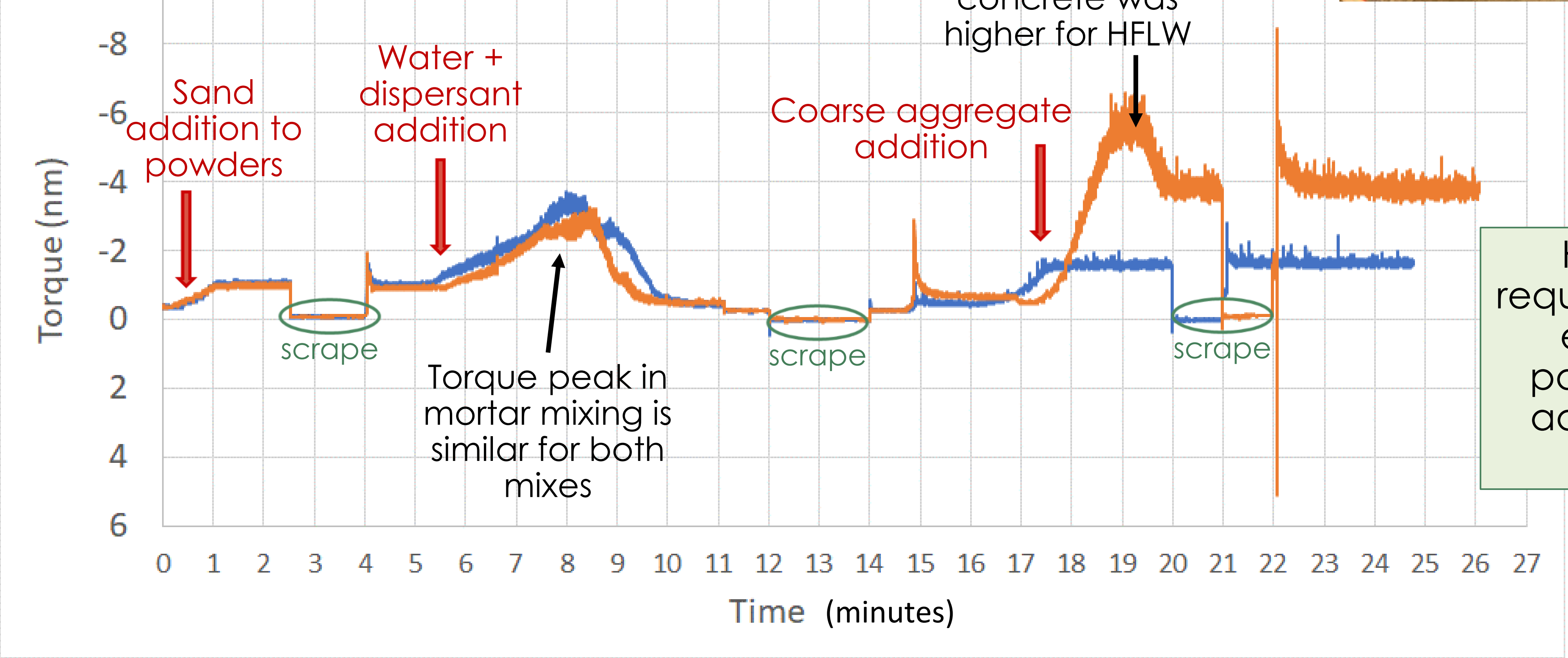




Modeling: Theoretical Concrete Designs

Design	**Reference**	**1**	**2**	**3**	**4**	**5**	**6**	**7**	**8**	**9**	**10**	**11**	**12**	**13**	**14**	**15**	**16**	**17**
Type IL cement (kg/m^3)	**427.1**	282.3	213.3	213.3	213.3	213.3	242.4	242.4	242.4	213.6	213.6	213.6	213.6	213.6	213.6	213.6	213.6	213.6
Filler 1 (kg/m^3)	**0.0**	281.7	213.8	213.8	213.8	213.8	243.5	130.2	186.9	213.8	213.8	213.8	213.8	213.8	185.5	157.1	185.5	157.1
Filler 2 (kg/m^3)	**0.0**	0.0	0.0	0.0	0.0	0.0	0.0	116.8	58.4	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0
Pozzolan (kg/m^3)	**0.0**	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	25.8	51.5	0.0	0.0
Fly Ash (kg/m^3)	**0.0**	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	0.0	25.8	51.5
Coarse aggregate (kg/m^3)	**1002.5**	1002.5	1075.0	1014.1	869.3	1159.0	1089.5	1089.5	1089.5	1008.3	927.2	869.3	1014.1	970.7	970.7	970.7	970.7	970.7
Sand (kg/m^3)	**735.9**	735.9	793.2	849.1	982.4	715.9	726.5	726.5	726.5	764.9	839.5	892.8	759.6	799.6	799.6	799.6	799.6	799.6
Water (kg/m^3)	**171.1**	118.9	118.9	118.9	118.9	118.9	118.9	118.9	118.9	131.2	131.2	131.2	131.2	131.2	131.2	131.2	131.2	131.2
PCE-based HRWR 1 (kg/m^3)	**1.07**	3.570	3.570	3.570	3.570	3.570	3.570	3.570	3.570									
Non-Cl accelerator (kg/m^3)		1.890	1.890	1.890	1.890	1.890	1.890	1.890	1.890	6.493	6.493	6.493	6.493	6.493	6.493	6.493	6.493	6.493
Air entraining agent (kg/m^3)										0.079	0.079	0.079	0.079	0.079	0.079	0.079	0.079	0.079
VMA (kg/m^3)										0.157	0.157	0.157	0.157	0.157	0.157	0.157	0.157	0.157
Nucleation agent + PCE (kg/m^3)										2.729	2.729	2.729	2.729	2.729	2.729	2.729	2.729	2.729
PCE-based HRWR 2 (kg/m^3)										4.082	4.082	4.082	4.082	4.082	4.082	4.082	4.082	4.082
Cost ($/yd^3)	**206.75**	222.72	204.51	204.97	206.06	203.88	211.95	201.10	206.52	210.31	210.92	211.36	210.27	210.59	206.36	202.14	206.36	202.14
Cost (% of Reference)	**100.0**	107.7	98.9	99.1	99.7	98.6	102.5	97.3	99.9	101.7	102.0	102.2	101.7	101.9	99.8	97.8	99.8	97.8
% CO_2	**15.5**	9.9	7.5	7.5	7.6	7.5	8.5	8.5	8.5	7.8	7.8	7.8	7.8	7.8	7.8	7.8	7.8	7.8
Vol surf area, $\phi<100\mu m$ (m^2/cm^3)	**4.261**	5.000	5.017	5.018	5.019	5.017	5.010	4.367	4.705	5.014	5.015	5.015	5.014	5.015	4.627	4.239	4.627	4.239
Concentration of fines, $\phi<100\mu m$ (%)	**44.96**	61.75	55.05	55.05	55.05	55.05	58.20	55.70	56.98	52.62	52.62	52.61	52.62	52.62	49.01	45.41	49.01	45.41
Packing porosity (%)	**10.73**	17.10	17.10	17.10	17.10	17.10	17.11	14.19	15.81	17.09	17.09	17.09	17.09	17.09	16.36	15.58	16.82	16.55
Interparticle Separation, IPS (μm)	**0.518**	0.165	0.243	0.243	0.243	0.243	0.204	0.289	0.241	0.277	0.277	0.277	0.277	0.277	0.365	0.480	0.362	0.474
Paste volume (%)	**37.88**	37.88	33.24	33.24	33.24	33.24	35.23	33.63	34.43	36.59	36.59	36.59	36.59	36.59	36.59	36.59	36.59	36.59
Vol surf area $\phi>100\mu m$ (m^2/cm^3)	**0.0056**	0.006	0.006	0.006	0.007	0.005	0.005	0.055	0.031	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006	0.006
Conc. coarse particles, $\phi>100\mu m$ (%)	**62.12**	62.12	66.76	66.76	66.76	66.76	64.77	66.37	65.57	63.41	63.41	63.41	63.41	63.41	63.41	63.41	63.41	63.41
Packing porosity (%)	**21.07**	21.07	21.03	19.89	19.05	22.41	21.84	21.39	21.62	20.79	19.11	18.62	20.90	20.05	20.05	20.05	20.05	20.05
Max Paste Thickness, MPT (μm)	**123.25**	123.25	83.05	84.63	78.84	81.48	99.42	8.47	16.26	111.44	111.81	108.53	111.46	111.48	111.48	111.48	111.48	111.48
w/c ratio	**0.401**	0.421	0.558	0.558	0.558	0.558	0.491	0.491	0.491	0.614	0.614	0.614	0.614	0.614	0.614	0.614	0.614	0.614
w/fines ratio	**0.401**	0.211	0.279	0.279	0.279	0.279	0.245	0.243	0.244	0.307	0.307	0.307	0.307	0.307	0.309	0.311	0.309	0.311

Concrete Rheometry During Mixing



Sand addition to powders

Water + dispersant addition

Torque peak in mortar mixing is similar for both mixes

Coarse aggregate addition

Torque to mix concrete was higher for HFLW

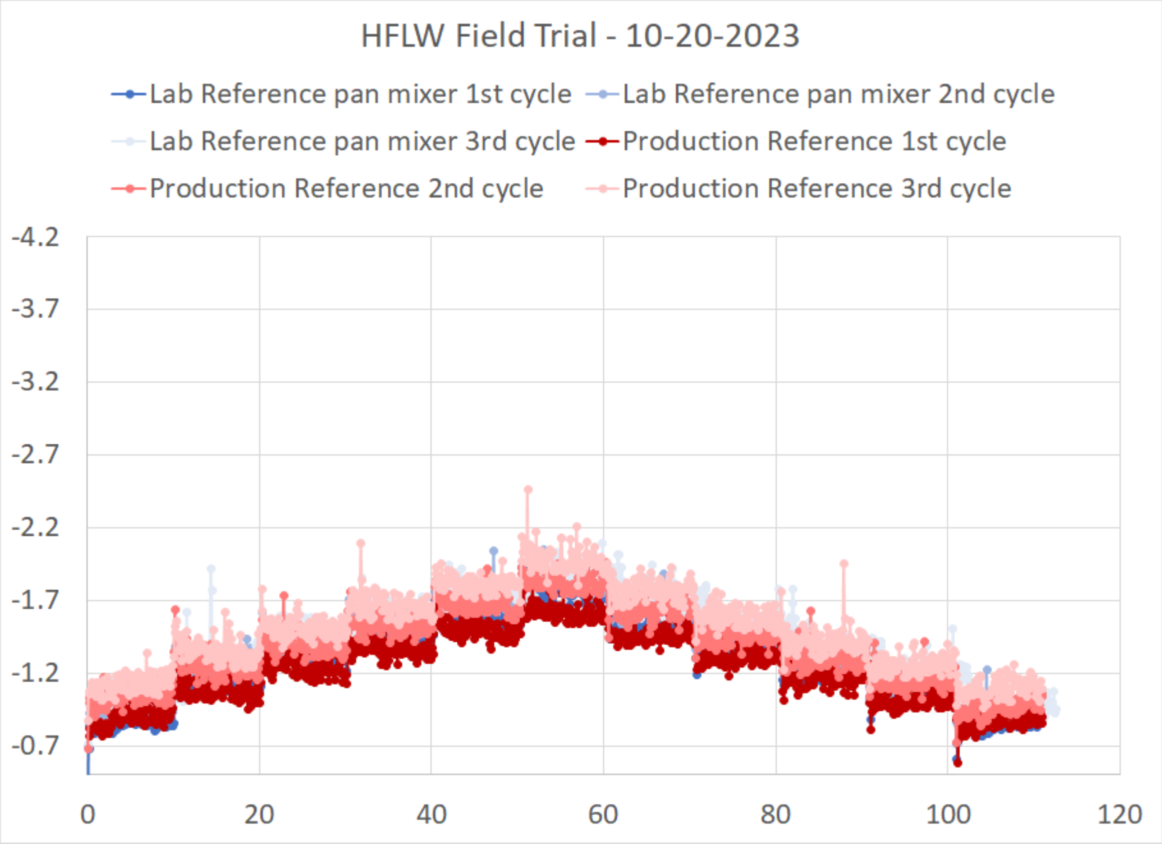
HFLW concrete required higher mixing energy, but this parameter can be adjusted to match reference.

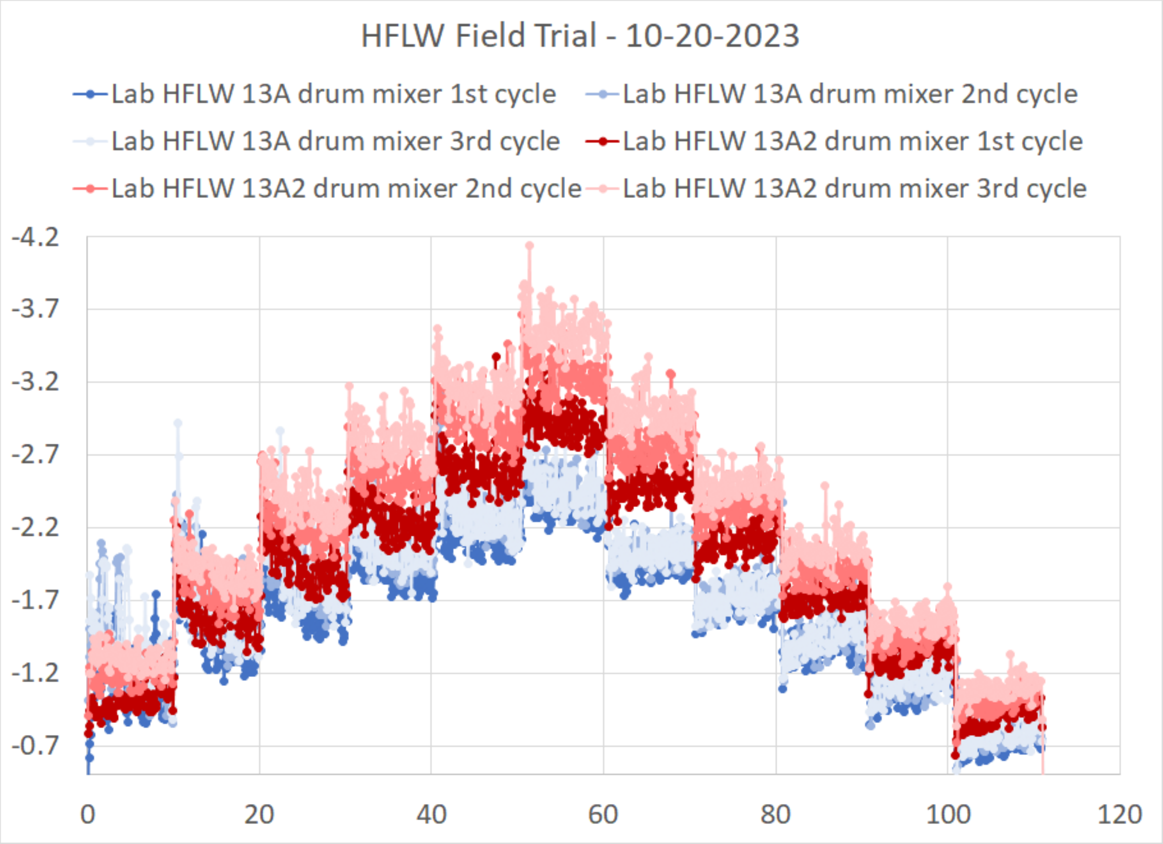
Preliminary Concrete Lab Testing Results

Parameter	Δ from lab reference
Type IL cement	- 51%
Limestone filler	Added 50%wt fines
Water	- 23%
PCE-based dispersant	+ 3x
Non-chloride accelerator	Manufacturer-recommended dose
w/c ratio	0.40 → 0.57
w/cm ratio	0.40 → 0.28
Cost	- 7%
Spread	SCC
Unit weight	+ 3%
Initial set time (UPV)	+ 6% (18 min)
Final set time (UPV)	+ 2% (6 min)
Compressive strength	Similar to higher at 12h and 24h

Lab-scale Test at Partner Precast Concrete Producer

- 2 reference mixes (lab-prepared, industrially-prepared in 5yd^3 pan mixer), 4 HFLW mixes
- Highlights:
  - – HFLW mixes are SCC, with 20hr and 7d compressive strength within spec
  - – Need further refinement - there is room to reduce water and dispersant.
  - – Cost of HFLW mixtures within ± 3% of reference.





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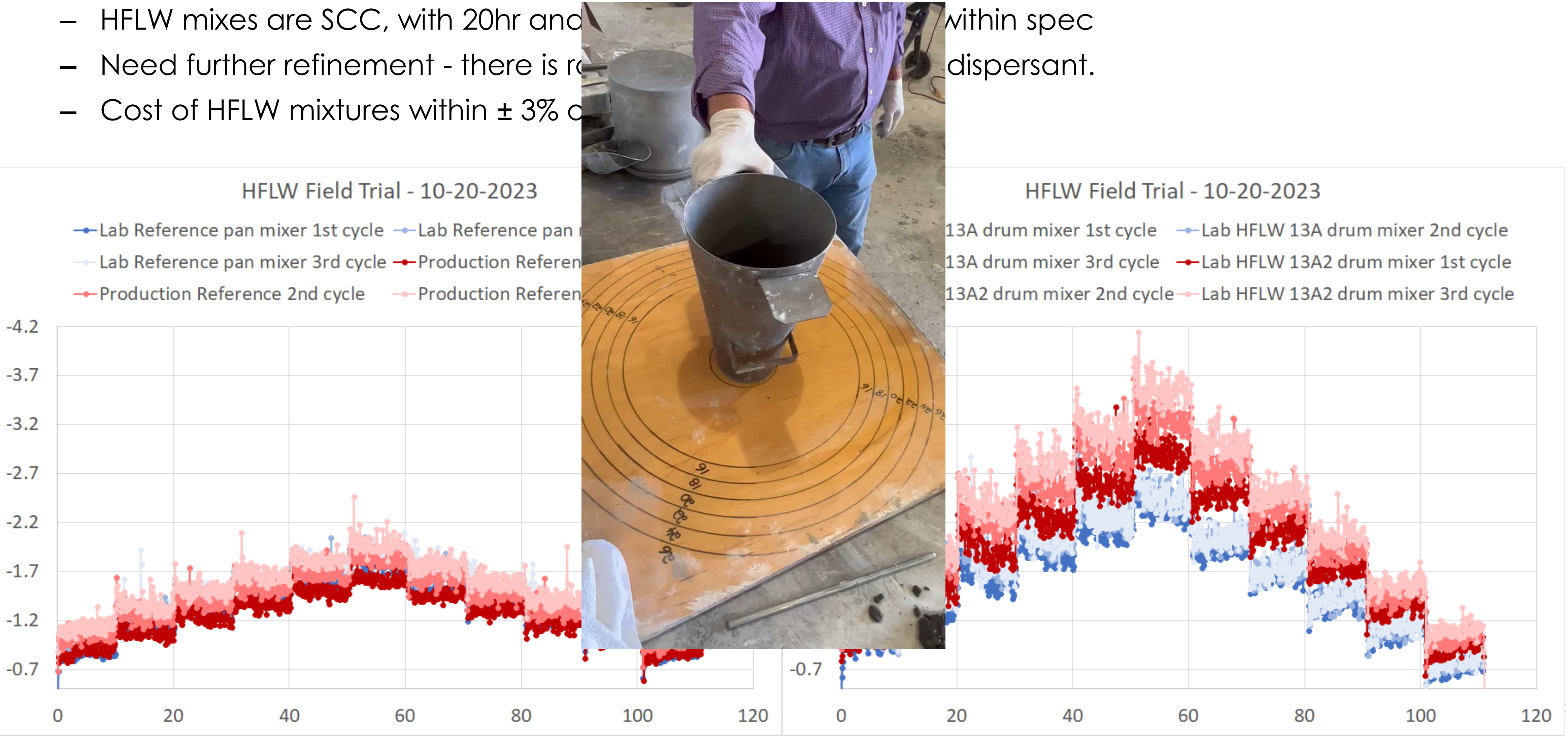

Concrete Lab Testing Results

Parameter	Δ from lab reference	Δ from lab reference at plant
Type IL cement	- 51%	- 51%
Limestone filler	Added 50%wt fines	Added 50%wt fines
Water	- 30%	-23%
PCE-based dispersant	+ 3x	+3.8x (admixture change)
Non-chloride accelerator	Manufacturer-recommended dose	Manufacturer-recommended dose
w/c ratio	0.40 → 0.57	0.40 → 0.61
w/cm ratio	0.40 → 0.28	0.40 → 0.31
Cost	- 7%	+1.7%
Spread	SCC	SCC
Unit weight	+ 3%	n/a
Initial set time (UPV)	+ 6% (18 min)	n/a
Final set time (UPV)	+ 2% (6 min)	n/a
Compressive strength	Similar to higher at 12h and 24h	Reference: 4793 psi @ 20 hrs HFLW: 4210 psi @ 17.5 hrs
		+32% at 7 days

Conclusions

- Particle packing and mobility models show potential to enable design of low carbon HFLW concrete mixtures using known materials and achieving:
  - **50% less Type IL cement**
  - **Similar setting times**
  - **Similar early mechanical performance**
  - **Similar cost**
- Scale-up effort is on-going at one partner prestressed concrete producer. Target is to scale up the technology in three producers by Sept. 2025.
- Concrete rheometer used at precast plants to benchmark the rheological properties of reference concrete as industrially produced. Focus is then to adjust HFLW concrete for similar rheological behavior.
- Precast producers will evaluate HFLW concrete performance for iterative design adjustment process as needed.
- LCA is on-going.

Questions?

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