Reducing Clinker Content and Carbon Footprint of Concrete Using SCMs, Limestone Cement, and Optimized Aggregate Gradations

Doug Hooton
Portland cement is the primary binder in Concrete

- Portland Cement is manufactured from limestone and shale rocks that have been fired at 1450 C to form a synthetic rock called clinker. This clinker is then crushed to a powder.
- When limestone is heated, it gives off CO$_2$.

\[ \text{CaCO}_3 \rightarrow \text{CaO} + \text{CO}_2 \]

- This reaction is unavoidable in the manufacture of cement clinker.
- So to reduce CO$_2$ the clinker fraction of cement has to be reduced.
Manufacture of Portland Cement

- Contributes up to 5% of global CO\(_2\) emissions
- Contributes up to 2% of global energy use
- For every tonne of cement produced:
  - 0.8 – 1.0 t of CO\(_2\) produced
  - 1,700 kWh of energy consumed/t
  - 1.5 t of raw material required
  - 3,300,000 t cement produced globally in 2010
- Cement is the most expensive concrete material component and can account for up to 60% of the total materials cost even though it is only approx. 10 – 15% by mass
- The cement paste fraction usually is 25% to 30% of the total volume of concrete
But cement is only one component of concrete

- ~90% of carbon footprint of concrete is from portland cement clinker (assuming portland cement is used as the sole cementing material)
There is no single right answer to reducing clinker content of concrete

- Optimization of combined aggregate gradations.
- Use of water reducing admixtures.
- Use of portland-limestone cements (PLC)
- Use of SCMs
- All can be done simultaneously
Optimizing Concrete Mixtures by Use of Supplementary Cementitious Materials (SCMs) and Portland-Limestone Cements (PLCs)

SCMs → PLC

<table>
<thead>
<tr>
<th>Portland cement type</th>
<th>Portland-Limestone cement type</th>
<th>Blended hydraulic cement type</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type I (GU)</td>
<td>Type IL (GUL)</td>
<td>Type IT (GUL_b)</td>
</tr>
</tbody>
</table>
Two approaches for reducing the carbon footprint of concrete

1. Reduce the clinker content of the cementitious binder
2. Reduce the total binder content of concrete mixtures.

For the first, combinations of supplementary cementitious materials can be combined with Type IL cements while still attaining early-age strength development with at least a 40% reduction in clinker content.

For the second, optimizing aggregate gradations with at least three size fractions can result in savings of up to 15% of the required cementitious materials content while also reducing concrete permeability and shrinkage.
More Cement is Not Always Better!

- At a fixed W/CM, more cement raises the unit water content of the mix and makes it more porous and more permeable.
- High cement contents can also lead to higher thermal stresses and increased shrinkage, making the concrete more vulnerable to cracking.
- Chemical admixtures can be used to obtain workable concretes at lower water (and cement) contents.
- Optimized aggregate gradations will also reduce water demand.
1. Increasing Aggregate Content

• Having to meet current ASTM, CSA and DOT specifications for meeting separate fine and coarse aggregate gradations can result in large portions of quarried and crushed stone being wasted only due to sieve sizes.
There is typically a gap when individual fine & coarse aggregates meet their individual grading envelopes.
Optimizing Combined Aggregate Gradation and using Microfine Fillers

**Typical Mix**
*Gap-graded*
- Gap-graded; lack of intermediate particles
- No microfine fillers; lack of \(<75\mu m\) particles
- ↑ void content
- ↑ paste fraction required

**Optimal Mix**
*Well-graded*
- Well-graded; plenty of intermediate particles
- Microfine fillers; plenty of \(<75\mu m\) particles
- ↓ void content
- ↓ paste fraction required
A well-graded combined aggregate blend can be achieved by using optimization techniques, or by adding low value or wasted coarse aggregate material of finer sieve sizes (1-5 mm).
Existing Optimization Techniques: Sieve Analysis

8-18 Distribution

Coarseness Factor (CF) Chart

Optimal for 12.5mm or finer

Too Fine

Gap-Graded

Well-Graded

Optimal

Too Coarse

% of Coarse over (Coarse + Intermediate) Aggregate

% of Fine Aggregate

Sieve Size (mm)

% Retained (by mass)
Blue lines show existing fine and coarse envelopes with deficiency between 2 & 5 mm
Red lines show smoother combined envelope
Optimized total aggregate gradation is now allowed to provide the opportunity to improve concrete performance, sustainability and economy by optimizing the aggregate envelope for the whole mix and not the individual components.

- Combination must include 3 or more separate components
- Material from all aggregate sources passing the 5 mm sieve shall be tested in the proportions to be used in the concrete mixture and the blend tested as a fine aggregate to show compliance with requirements.
- Material retained on the 5 mm sieve shall be tested to show compliance with coarse aggregate requirements.
Workability of optimized gradation mixes also needs to be evaluated.
### Comparison of concretes with and without 31% (0.3 to 5 mm) limestone screenings: w/cm = 0.39, normal water reducer and air entrained to 5-8% air. (Anson-Cartwright, PhD)

<table>
<thead>
<tr>
<th></th>
<th>Type I +25% Slag</th>
<th>Type I + 25% Slag</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total Cementitious Content (kg/m³)</td>
<td>360</td>
<td>330</td>
</tr>
<tr>
<td>Cement Type</td>
<td>Type I</td>
<td>Type I +25% Slag</td>
</tr>
<tr>
<td>Limestone Screenings</td>
<td>No</td>
<td>Yes</td>
</tr>
<tr>
<td>MRWR Dose for 80-120 mm slump (mL/100 kg)</td>
<td>935</td>
<td>950</td>
</tr>
<tr>
<td>28 day Strength (MPa)</td>
<td>57.8</td>
<td>69.2</td>
</tr>
<tr>
<td>28 day drying shrinkage</td>
<td>0.033%</td>
<td>0.025%</td>
</tr>
<tr>
<td>ASTM C1202 (coulombs @ 56 days)</td>
<td>900</td>
<td>640</td>
</tr>
</tbody>
</table>
Proper Gradation of Aggregates can save up to 15% of cement (Anson-Cartwright & Hooton 2011)

Well-graded aggregate

Low permeability

Poorly-graded aggregate

High permeability

Same w/c of paste fraction
For individual and combined aggregate materials:

- Gradation (including nominal maximum size and size distribution)
  - The more gap-graded and coarser the combined aggregate gradation, the higher the cement paste content required

- Shape (spherical, cubical, flat, or elongated)
  - The more cubical / spherical the particles, less cement paste is required
  - Cubical is best for packing and spherical best for workability

- Angularity (angular or rounded)
  - The more rounded the particles, less cement paste is required

- Surface Texture (rough or smooth)
  - The smoother the particles, the less cement paste (effectively less water demand) required
Cementitious contents can be reduced

- 16% reductions in 50 MPa (7250 psi) bridge deck mixes were obtained (465 to 390 kg/m³) while meeting 1000 coulomb limit @56d. (775 → 650 pcy)

- 8% reductions in 35MPa mixes were obtained (360 to 330 kg/m³) while still meeting a 1500 coulomb limit @ 56d. (600 → 550 pcy)

- This was with use of an intermediate size C. Agg. to fill the gap between fine and coarse agg. fractions
While supplementary cementitious materials (SCMs) such as slag and fly ash can be used to reduce the clinker content of concrete, another initiative is to intergrind the cement clinker with raw limestone.

SCMs and limestone also work well when used together, so limestone cements do not require reducing SCM levels.

This directly reduces point-source CO$_2$ emissions at cement plants by $\sim$10%.

ASTM C595 Type IL allows up to 15% limestone as does CSA Type GUL
Sustainable Development

Use of portland-limestone cements
• Reduces CO$_2$ emissions (by 10% over current portland cements)
• Reduces impact on natural resources, since 46% less limestone is used than when it is processed into clinker
• Reduces energy consumption (coal)
Performance Requirements

• In ASTM C595 and CSA A3000, the setting times and strength development limits are the same for Type IL (GUL in Canada) as for portland cements.

• Type IL cements typically perform better with SCMs than Type I in terms of strength and permeability. This is due to formation of calcium carbo-aluminates.
**Strengths of Air-entrained Concretes cured at 23 °C with limestone and SCMs**

<table>
<thead>
<tr>
<th>Mix Identification (all 400 kg/m³ (666 pcy mixes))</th>
<th>% clinker in binder</th>
<th>w/cm</th>
<th>Compressive Strength (MPa)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td>7 day</td>
</tr>
<tr>
<td><strong>Type I (GU) Control</strong></td>
<td>89*</td>
<td>0.40</td>
<td>39.3</td>
</tr>
<tr>
<td><strong>GU + 40% Slag</strong></td>
<td>53</td>
<td>0.40</td>
<td>32.8</td>
</tr>
<tr>
<td><strong>Type IL (9%L) + 40% Slag</strong></td>
<td>50</td>
<td>0.40</td>
<td>36.1</td>
</tr>
<tr>
<td><strong>Type IL (9%L) + 50% Slag</strong></td>
<td>41</td>
<td>0.40</td>
<td>34.6</td>
</tr>
<tr>
<td><strong>Type IL (15%L) + 40% Slag</strong></td>
<td>46</td>
<td>0.40</td>
<td>37.1</td>
</tr>
<tr>
<td><strong>Type IL (15%L) + 50% Slag</strong></td>
<td>38</td>
<td>0.40</td>
<td>36.3</td>
</tr>
<tr>
<td><strong>Type IL (15%L) + 6% Silica Fume + 25% Slag</strong></td>
<td>53</td>
<td>0.40</td>
<td>46.0</td>
</tr>
</tbody>
</table>

* 3.5% limestone and 8% gypsum

U of Toronto Field site mixes
### RCPT of Air-entrained Concretes cured at 23 °C with limestone and SCMs

<table>
<thead>
<tr>
<th>Mix Identification (all 400 kg/m³ (666 pcy mixes))</th>
<th>% clinker in binder</th>
<th>w/cm</th>
<th>Rapid Chloride Permeability (Coulombs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Type I Control</td>
<td>89</td>
<td>0.40</td>
<td>2384</td>
</tr>
<tr>
<td>Type I + 40% Slag</td>
<td>53</td>
<td>0.40</td>
<td>800</td>
</tr>
<tr>
<td>Type IL 9% + 40% Slag</td>
<td>50</td>
<td>0.40</td>
<td>867</td>
</tr>
<tr>
<td>Type IL 9% + 50% Slag</td>
<td>41</td>
<td>0.40</td>
<td>625</td>
</tr>
<tr>
<td>Type IL 15% + 40% Slag</td>
<td>46</td>
<td>0.40</td>
<td>749</td>
</tr>
<tr>
<td>Type IL 15% + 50% Slag</td>
<td>38</td>
<td>0.40</td>
<td>525</td>
</tr>
<tr>
<td>Type IL 15% + 6% Silica Fume + 25% Slag</td>
<td>53</td>
<td>0.40</td>
<td>357</td>
</tr>
</tbody>
</table>
Example of MTO Highway Field Trials

a) Nov. 4, 2009

- Dufferin Construction Barrier Wall Test
  sections 23m$^3$ of PLC+15% Slag vs GU+15% Slag (CM = 355 kg/m$^3$)
- QEW in Burlington
- First MTO trial of PLC
- Testing performed by Dufferin and University of Toronto, with scaling slabs also tested by MTO.
23 m³ of each mix placed, 30 MPa, 60-100 mm (2.5-4 in.) slump
### Nov. 2009 Barrier Wall

<table>
<thead>
<tr>
<th>2009 Barrier Wall</th>
<th>PC +25% SLAG</th>
<th>PLC + 25% SLAG</th>
</tr>
</thead>
<tbody>
<tr>
<td>Shrinkage (28d)</td>
<td>0.038%</td>
<td>0.038%</td>
</tr>
<tr>
<td>Strength (MPa)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>1</td>
<td>9.5</td>
<td>10.3</td>
</tr>
<tr>
<td>3</td>
<td>19.3</td>
<td>19.4</td>
</tr>
<tr>
<td>7</td>
<td>25.6</td>
<td>26.8</td>
</tr>
<tr>
<td>28</td>
<td>36.9</td>
<td>37.9</td>
</tr>
<tr>
<td>56</td>
<td>38.9</td>
<td>38.0</td>
</tr>
<tr>
<td>91</td>
<td>40.7</td>
<td>40.2</td>
</tr>
<tr>
<td>Freeze/Thaw Durability</td>
<td>94%</td>
<td>94%</td>
</tr>
<tr>
<td>MTO LS-412 Scaling</td>
<td>0.24 kg/m²</td>
<td>0.24 kg/m²</td>
</tr>
<tr>
<td>RCP (Coulombs)</td>
<td></td>
<td></td>
</tr>
<tr>
<td>28 days</td>
<td>2070</td>
<td>1490</td>
</tr>
<tr>
<td>56 days</td>
<td>1930</td>
<td>1340</td>
</tr>
</tbody>
</table>
Nov. 2009 Barrier Wall Scaling Tests

<table>
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<tr>
<th>2009 Barrier Wall</th>
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<th>PLC + 25% SLAG</th>
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<tbody>
<tr>
<td>MTO LS-412 Scaling</td>
<td>0.35 kg/m²</td>
<td>0.51 kg/m²</td>
</tr>
<tr>
<td>UofT LS-412 Scaling</td>
<td>0.24 kg/m²</td>
<td>0.24 kg/m²</td>
</tr>
</tbody>
</table>

MTO scaling limit is 0.8 kg/m²
Since 2009

- Several more MTO field trials in 2010-2012 showed benefit of using PLC
- Increasing use of Portland-limestone cements with SCMs in pavements and industrial/commercial applications
Concrete Optimization

**Cause**

*Reduced paste fraction by:*
- Optimization of Combined Aggregate Gradation
- Use of water reducing admixtures

*Reduced Portland cement content by:*
- Addition of Interground Limestone
- Addition of Supplementary Cementitious Materials

**Effect**

*Performance:*
- ↑ Strength

*Durability:*
- ↓ Permeability
- ↓ Shrinkage

*Sustainability and Cost:*
- ↓ Cement content
Possible Cumulative Reduction in Cement Contents (from 12% to 3% by volume)

From 380 kg/m³ to 95 kg/m³ (633pcf to 158 pcf)

Anson-Cartwright & Hooton 2011
Two approaches for reducing the carbon footprint of concrete are to,

1. Reduce the clinker content of the cementitious binder: Combinations of supplementary cementitious materials can be combined with Type IL cements while still attaining early-age strength development with at least a 40% reduction in clinker content.

2. Reduce the total binder content:
   (a) Optimizing aggregate gradations with at least three components can result in savings of up to 15% of the required cementitious materials content while also reducing concrete permeability and shrinkage.
   (b) Use water-reducing admixtures.
The carbon footprint of concrete can be reduced by fairly simple changes to materials and mix proportions.

When aggregate gradations are optimized, and the binder contains both SCMs and limestone, the clinker content of concrete can be reduced by a factor of up to 60%.

Since 90% of the carbon footprint of concrete is from cement, these measures would reduce the footprint concrete by as much as 60%.