Physical and chemical characteristics of natural limestone fillers: mix properties and packing density

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TC-SCM WORKSHOP, CYPRUS, 29-30 MARCH 2012

Outline

- Introduction
- Materials
- Physico-chemical characteristics
- Packing properties
- Conclusions
Specific requirements for fresh SCC: high workability and good resistance to segregation.

Amount of coarse aggregate reduced and replaced by fine material.

In Belgium, local available materials = limestone fillers.

Suitability of these fillers for use in SCC or conventional concrete?

Production process of limestone fillers

Aggregate and lime production industry (quarrying operations)
Introduction

Production process of limestone fillers

Ornamental stones industry (sawing operations)

Materials

Ordinary Portland Cement (PC) CEM I 42,5 R HES
Six limestone fillers collected in Belgium (F1 to F6)

<table>
<thead>
<tr>
<th>Limestone filler reference</th>
<th>Production process</th>
<th>Industrial sector</th>
</tr>
</thead>
<tbody>
<tr>
<td>F1</td>
<td>Dry process</td>
<td>Lime</td>
</tr>
<tr>
<td>F2</td>
<td>Crushing</td>
<td>Aggregates</td>
</tr>
<tr>
<td>F3</td>
<td>Drying / crushing</td>
<td>Aggregates</td>
</tr>
<tr>
<td>F4</td>
<td>Sawing</td>
<td>Ornamental stones</td>
</tr>
<tr>
<td>F5</td>
<td>Washing</td>
<td>Aggregates</td>
</tr>
<tr>
<td>F6</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>
## Physical characterization (Laser diffraction)

![Laser diffraction graph](image)

<table>
<thead>
<tr>
<th></th>
<th>F1</th>
<th>F2</th>
<th>F3</th>
<th>F4</th>
<th>F5</th>
<th>F6</th>
<th>PC</th>
</tr>
</thead>
<tbody>
<tr>
<td>$d_0$ (µm)</td>
<td>13.6</td>
<td>9.4</td>
<td>8.8</td>
<td>7.1</td>
<td>9.0</td>
<td>14.8</td>
<td>16.6</td>
</tr>
<tr>
<td>$S_{BET}$ (m$^2$/g)</td>
<td>1.3</td>
<td>1.2</td>
<td>5.5</td>
<td>4.0</td>
<td>5.7</td>
<td>3.7</td>
<td>—</td>
</tr>
</tbody>
</table>

## Physico-chemical characteristics

### Mineralogical and chemical characterization

<table>
<thead>
<tr>
<th></th>
<th>F1</th>
<th>F2</th>
<th>F3</th>
<th>F4</th>
<th>F5</th>
<th>F6</th>
</tr>
</thead>
<tbody>
<tr>
<td>Calcite CaCO$_3$ [%]</td>
<td>99.5</td>
<td>99.5</td>
<td>82.0</td>
<td>94.5</td>
<td>86.0</td>
<td>75.0</td>
</tr>
<tr>
<td>Quartz SiO$_2$ [%]</td>
<td>0.0</td>
<td>0.0</td>
<td>15.5</td>
<td>1.8</td>
<td>6.5</td>
<td>2.0</td>
</tr>
<tr>
<td>Dolomite Ca(Mg,Fe)(CO$_3$)$_2$ [%]</td>
<td>0.5</td>
<td>0.5</td>
<td>2.5</td>
<td>3.7</td>
<td>7.5</td>
<td>23.0</td>
</tr>
<tr>
<td>Methylene Blue Adsorption MBA [g/kg filler]</td>
<td>0.7</td>
<td>0.7</td>
<td>4.0</td>
<td>1.3</td>
<td>5.0</td>
<td>3.3</td>
</tr>
</tbody>
</table>

- Fillers coming from lime production (F1, F2) and ornamental stones sawing (F4): high CaCO$_3$ content.
- Fillers produced in limestone quarries (F3, F6): large amounts of impurities.
**Physico-chemical characteristics**

**Bêta-P: spread measurement for different W/P**

\[
\tau_p = \left( \frac{D_{m1} + D_{m2}}{2D_0} \right)^2 - 1
\]

\[
y = 0.049x + 1.0734 \\
R^2 = 0.9551
\]

**Smooth Paste test (Legrand, 1971)**

Modification of the paste appearance

\[V_{E} \rightarrow \]

... correlated with threshold value of dilantancy (rheological behaviour)
Physico-chemical characteristics

Relationship between mortar flowability and MBA or $b_p$ of limestone fillers

![Graph showing the relationship between mortar flowability and MBA or $b_p$.]

Physico-chemical characteristics

Relationship between $S_{S,BET}$ and MBA

![Graph showing the relationship between $S_{S,BET}$ and MBA.]

\[ R^2 = 0.8412 \]
### Physico-chemical characteristics

#### Materials

<table>
<thead>
<tr>
<th>Material</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>CEM I 42.5</td>
<td></td>
</tr>
<tr>
<td>CEM I 52.5</td>
<td></td>
</tr>
<tr>
<td>Limestone Filler</td>
<td></td>
</tr>
<tr>
<td>Standard sand EN196-1:2005 (0~2 mm)</td>
<td></td>
</tr>
</tbody>
</table>

#### Size and shape characterization

**Equipments**

- **OCCHO 500Nano** (0.5 µm~2 mm)
- Vacuum dispersion
- Image analysis
**Physico-chemical characteristics**

### Particle size

**Traditional equivalent volume (area) diameter**

- Maximum inscribed diameter

![Graph showing particle size distribution](image)

**PSD:**

- Passing fraction [%]

- Size [µm]

#### Physical and chemical characteristics

**Particle shape**

- **Elongation**
  
  \[ \text{Elongation} = 1 - \frac{b}{a} \]

- **Circularity**
  
  \[ \text{Circularity} = \sqrt{\frac{4\pi A}{P^2}} \]

![Images of particle shapes](image)

**Particles >6 µm (>500 pixels/particle):**

- CEM I 52.5
- CEM I 42.5

- Holzer et al., 2010 by pCT & FIB-NT

**Average inner diameter (µm):**

- LF
- CEM I 52.5
- CEM I 42.5

- Elongation LF
- Elongation CEM I 52.5
- Elongation CEM I 42.5
**Physico-chemical characteristics**

### Particle shape

**Roundness**

\[ \text{Roundness} = \frac{4A}{\pi F_{\text{max}}} \]

**Solidity**

\[ \text{Solidity} = \frac{A}{A_c} \]

### Bluntness

Bluntness describes the maturity of the particle in the abrasion process.

\[ \text{Bluntness} = \frac{1}{\sqrt{\overline{V}} - 1} \]

In which:

\[ \overline{V} = \frac{1}{N} \sum_{i=1}^{N} \left(1 + \frac{r_{\text{max}}}{r_i}\right)^3 \]

**Krumbein’s chart**

Calypter tools

**Dimension value**

- **Bluntness LF**
- **Bluntness CEM 52.5**
- **Bluntness CEM 42.5**

**Average inner diameter [µm]**

- **Solidity LF**
- **Solidity CEM 52.5**
- **Solidity CEM 42.5**

- **Roundness LF**
- **Roundness CEM 52.5**
- **Roundness CEM 42.5**
**Packing properties**

Dry packing (direct) methods, e.g. BS 812:Part 2:1995
- For aggregate
- For fillers
> Influences of inter-particle forces?
> Standard of compaction level?

Wet packing (indirect) methods:
- Standard consistence test, BS EN 196:part 3, 1995
- the wet packing method (Wong & Kwan, 2008)

**The wet packing method** (Wong & Kwan, 2008)

Packing density:
\[
\phi = \frac{V_b}{V} = \frac{M_b}{V \rho_s} = \frac{M}{V(u \rho_a + \rho_s)}
\]

The voids ratio (\(u\)):
\[
u = \frac{V - V_b}{V_b} = \frac{1}{\phi} - 1
\]
Packing properties

Experiments

The wet packing method (Wong & Kwan, 2008)

The dry packing method

- Influences of inter-particle forces?
- Standard of compaction level?

Compaction cylinder

Concrete vibration table

Packing tests

The dry packing method

<table>
<thead>
<tr>
<th>Vibration time [s]</th>
<th>Packing density (PD) [-]</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.3</td>
</tr>
<tr>
<td>30</td>
<td>0.4</td>
</tr>
<tr>
<td>60</td>
<td>0.5</td>
</tr>
<tr>
<td>90</td>
<td>0.6</td>
</tr>
<tr>
<td>120</td>
<td>0.7</td>
</tr>
<tr>
<td>150</td>
<td>0.8</td>
</tr>
<tr>
<td>180</td>
<td>0.9</td>
</tr>
</tbody>
</table>

- PSD?
- Shape?
- Inter-particle forces?
Packing properties

The dry packing method

The wet packing method

Influence of entrapped air
Packing properties

Comparisons of the dry method and the wet packing method

Discussion on the wet packing method

Limitations in the wet packing method
- manual effects on M-V evaluation
- Superplasticizer

Cement hydration

Mixing efficiency (e.g. LF)
Packing properties

Results on blended cement (total replacement and coarse replacement)

![Graph showing packing density vs. volume fraction of LF]

Packing properties

Numerical simulations

- PSD simulations

![Graph showing sieve size vs. passing fraction]
Packing properties

Visualized models

Periodical boundaries:

- CEM I 42.5
  - 0.72
- CEM I 52.5
  - 0.68
- LF
  - 0.716

Rigid boundaries:

- CEM I 42.5
  - 0.674
- CEM I 52.5
  - 0.658
- LF
  - 0.695

Packing properties

Maximum packing density

- CEM I 42.5
- CEM I 52.5
- Limestone fillers (LF)
Packing properties

Surface to surface nearest neighboring distance (NND)

Surface-to-surface NND ($\mu m$)

Probability (-)

CEM I 42.5
CEM I 52.5
LF

Packing properties

Surface area density ($S_v$) and volume density ($V_v$) in ITZ

Distance to surface of aggregate ($\mu m$)

$S_v$, $V_v$ in CEM I 42.5, CEM I 52.5, LF
**Packing properties**

**Mechanical bounding capacities**

Meaning free spacing: \( \lambda = \frac{1 - V_v}{S_v} \)

A parameter proportional to global bonding capacity: \( \lambda^{-3} \)

[Stroeven & Stroeven 2001](#)

[Hu, 2004](#)

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**Permeability**

\( \kappa = \frac{(1 - V_v) \lambda^2}{32 + 16V_v} \)

[Carman 1939](#)

[Hu, 2004](#)
Conclusions

- The limestone fillers collected in Belgium differ from each other through their physico-chemical characteristics (impurities such as clay, quartz and dolomite).
- The water requirement of limestone fillers is mainly influenced by their clay content (indicated by high MBA and $S_{S,BET}$ values).
- Size and shape characteristics of LF and OPC can be identified by an advanced image analysis system.
- With a proper replacement of cement by LF, packing density of the mixture can be improved. Filler effect is significant as also illustrated by the numerical simulation.

Conclusions

Thank you for your attention