LC$^3$: A breakthrough technology to reduce CO$_2$ emissions from cementitious materials

Professor Karen Scrivener, FREng
We don't have much time left!

https://www.mcc-berlin.net/en/research/co2-budget.html
Drastic consequences!

North China Plain threatened by deadly heatwave due to climate change and irrigation

Suchul Kang & Elaith A.B. Eltahir

Guardian graphic. Source: Nature Communications
Two reports in recent years

Eco-efficient cements: Potential economically viable solutions for a low-CO$_2$ cement-based materials industry

A SUSTAINABLE FUTURE FOR THE EUROPEAN CEMENT AND CONCRETE INDUSTRY

Technology assessment for full decarbonisation of the industry by 2050

European perspective

Global perspective
Cement Based Materials: cannot be replaced by alternatives

Cementitious materials make up ~50% of everything we produce. In the light of this, CO$_2$ emissions of 5-10% very good
Concrete is an environmentally friendly material

<table>
<thead>
<tr>
<th>Material</th>
<th>MJ/kg</th>
<th>kgCO₂/kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cement</td>
<td>4.6</td>
<td>0.83</td>
</tr>
<tr>
<td><strong>Concrete</strong></td>
<td><strong>0.95</strong></td>
<td><strong>0.13</strong></td>
</tr>
<tr>
<td>Masonry</td>
<td>3.0</td>
<td>0.22</td>
</tr>
<tr>
<td>Wood</td>
<td>8.5</td>
<td>0.46</td>
</tr>
<tr>
<td>Wood: multilayer</td>
<td>15</td>
<td>0.81</td>
</tr>
<tr>
<td>Steel: Virgin</td>
<td>35</td>
<td>2.8</td>
</tr>
<tr>
<td>Steel: Recycled</td>
<td>9.5</td>
<td>0.43</td>
</tr>
<tr>
<td>Aluminium: virgin</td>
<td>218</td>
<td>11.46</td>
</tr>
<tr>
<td>Aluminium recycled</td>
<td>28.8</td>
<td>1.69</td>
</tr>
<tr>
<td>Glass fibre composites</td>
<td>100</td>
<td>8.1</td>
</tr>
<tr>
<td>Glass</td>
<td>15.7</td>
<td>0.85</td>
</tr>
</tbody>
</table>

ICE version 1.6a
Hammond G.P. and Jones C.I
2008 Proc Instn Civil Engineers
www.bath.ac.uk/mech-eng/sert/embodied/
Forecast growth

We need solutions for people in developing countries
How to meet this challenge sustainably

Solutions need to be:

➢ Practical,
  usable by unskilled workers

➢ Economically viable
What is available on earth?

- **Na<sub>2</sub>O**
  - Too soluble

- **K<sub>2</sub>O**
  - Too low mobility in alkaline solutions

- **Fe<sub>2</sub>O<sub>3</sub>**

- **MgO**

- **CaO**

- **SiO<sub>2</sub>**

- **Al<sub>2</sub>O<sub>3</sub>**
  - The most useful

30-year-old concrete

Slag cement blend
Hydraulic minerals in the system CaO-SiO$_2$-Al$_2$O$_3$

BUT, what sources of minerals are there which contain Al$_2$O$_3$ >> SiO$_2$?

Bauxite – localised, under increasing demand for Aluminium production, EXPENSIVE

Even if all current bauxite production diverted would still only replace 10-15% of current demand.

Less CaO > less CO$_2$
Portland based cement will continue to be dominant

- Incredible economy of scale
  - Clinker very low cost
- Raw materials abundant nearly everywhere
- Easily to manipulate open time
- Robust
Extending use of blended cements
Evolution of Clinker substitution

Clinker substitution most successful strategy to reduce CO₂

- Almost no progress in last 8 years
- Only 3 substitutes used in quantity
Availiability of SCMs

- silica fume
- waste glass
- Vegetable ashes
- Natural Pozzolan
- Slag
- Fly ash
- Portland cement
- limestone
- Calcined Clay

Classic SCMs – fly ash and slag are only around 15% of current cement production, will drop to < 10% in near future
There is no magic solution

- Blended with SCMs will be best solution for sustainable cements for foreseeable future

- Only material really potentially available in viable quantities is calcined clay.

- **Synergetic reaction** of calcined clay and limestone allows high levels of substitution:
  EPFL led LC$^3$ project supported by SDC. **Started 2013**
LC$^3$ project partners

- EPFL
- UCLV, Cuba; Fernando Martirena
- TARA, India; Soumen Maity
- IIT Delhi, India; Shashank Bishnoi
- IIT Madras, India; Ravindra Gettu, Manu Santhanam
- Sinoma, China; Sui Tongbo
What is LC³

LC³ is a family of cements, the figure refers to the clinker content.

- 50% less clinker
- 40% less CO₂
- Similar strength
- Better chloride resistance
- ASR resistant
Why can we get such high replacement levels

» Calcination of kaolinite at 700-850°C gives metakaolin: much more reactive than glassy SCMs
  - aluminium
  - silicon

» Synergetic reaction of Alumina in metakaolin with limestone to give space filling hydrates

![Graph showing X-ray diffraction patterns of OPC and LC^3-50](image)
What kinds of clay are suitable?
Three basic clay structures

**Kaolinite (1:1)**
- Aluminium
- Silicon

**Montmorillonite (2:1)**
(Smectites)
- Na⁺, Ca²⁺, H₂O

**Illite (Micas)**
(2:1)

“Metakaolin”, sold as high purity product for paper, ceramic, refractory industries
Requirements for purity, colour, etc, mean expensive 3-4x price cement

Clays containing metakaolin available as wastes
– over or under burden NOT agricultural soil
*Much much less expensive often available close to cement plants*
Over 50 clays studied from around the world

Different calcination conditions
Different compositions, impurities
Different physical properties

<table>
<thead>
<tr>
<th>% of calcined kaolinite in the calcined clay</th>
</tr>
</thead>
<tbody>
<tr>
<td>Quartz (0%)</td>
</tr>
<tr>
<td>17.0%</td>
</tr>
<tr>
<td>35.0%</td>
</tr>
<tr>
<td>8.9%</td>
</tr>
<tr>
<td>50.3%</td>
</tr>
<tr>
<td>66.2%</td>
</tr>
<tr>
<td>79.4%</td>
</tr>
<tr>
<td>95%</td>
</tr>
</tbody>
</table>

Pure kaolinite
Calcination Temperature window 700-850°

- **<600°** Not reactive
- **600-700°** Dehydroxilation complete but reactivity increases
- **850-1000°** Surface area reduced, reactivity decreases
- **>1000°** Crystalline phases formed, no reactivity
Benchmark test of clay strength

- Compressive strength EN 196-1 at 1, 3, 7, 28 and 90 d
- Linear increase of strength with the MK content of calcined clays
- Similar strength to PC for blends containing 40% of calcined kaolinite from 7d onwards
- At 28 and 90 days, little additional benefit >60%
- Minor impacts of fineness, specific surface and secondary phases

Calcined kaolinite content overwhelming parameter
Ideal kaolinite content

40-60%

Higher contents, possible to use more limestone

Even better economics and ecology

Lower contents can be enriched by separation

separated fine quartz can be sold as separate product
Availability of suitable clays, yellow pink and light green regions, and others
Suitable clays presently stockpiled as waste
Comparison of calcined kaolinitic clay, slag and fly ash

**Binary systems 70% clinker**
**Ternary systems, with limestone 50% clinker**
**Potential impact of LC³ technology**

<table>
<thead>
<tr>
<th></th>
<th>Global cement production</th>
<th>Clinker factor, global average</th>
<th>Global SCM volume</th>
<th>Global CO₂ reduction</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Billion tons/year</td>
<td>%</td>
<td>Billion tones/year</td>
<td>Million tones/year</td>
</tr>
<tr>
<td>2006</td>
<td>2.6</td>
<td>79</td>
<td>0.5</td>
<td></td>
</tr>
<tr>
<td>2050 (CSI study)</td>
<td>4.4</td>
<td>73</td>
<td>1.2</td>
<td>200</td>
</tr>
<tr>
<td>2050 (with LCC)</td>
<td>4.4</td>
<td>60</td>
<td>1.8</td>
<td>600</td>
</tr>
</tbody>
</table>

**IEA:** International Energy Agency **study for**

**CSI:** Cement Sustainability Initiative

**of WBCSD:** World Business Council for Sustainable Development

Global potential of LC³

\[ \Delta = 400 \text{ million tonnes per yr} \]

> whole of CO₂ emissions of France
LC³ has been produced and used in full scale trials

Uses existing technology
Rotary kiln
Flash calciner
etc
Cuba – 1st Industrial trial

Jan 2013: Clay sourcing Pontezuela (300 t)

March 2013: Clay calcination Siguane (110 t)

August 2013: Cement grinding (130 t)

Sept-Dec. 2013: Cement use in construction

Jan-July 2014: Evaluation of concrete made with LC3
Industrial block manufacture plant
Prefabrication plant Cuba
House built at Santa Clara, Cuba with LC3
INDIA: Calcination

Rotary kiln
Blending and grinding
Evaluation in building materials
Evaluation in building materials
Industrial production
KJS Concrete Pvt. Ltd., Dadri
Demonstration structure

Around 14 tonnes of CO$_2$ saved
Compared to existing solutions
Hollow Core Slabs
Key Advantages

• Chloride resistance
• ASR mitigation
Chloride ponding ASTM

Apparent diffusion coeffs.
Porosity characterization by MIP

➢ Significant refinement of porosity already at 3 days of hydration
Porosity characterization by MIP

- Significant refinement of porosity already at 3 days of hydration
Very dense microstructure

Strong pore refinement
Alkali silica reaction

Impact of alumina on aggregates

- PC Jobe 0.32 M
- LC3-50 (50%)Jobe 0.32M
- PC Jobe 1.6M
- LC3-50(95%)Jobe 0.32M
- PC Verz 0.32 M
- LC3-50 (50%)Verz 0.32M
- PC Verz PS

Expansion (%)

0.2
0.15
0.1
0.05
0

Time (d) in solution

10 210 410 610 810 1010 1210

No alumina

Alumina in solution

Chappex 2012
Perceived problems

• Workability
• Carbonation
• Colour
Limestone and calcined clay are both much softer than clinker.

With intergrinding, high blaine and clinker is likely to be underground. But situation can be improved by separate grinding or addition of calcined clay at separator.

However, the effect of limestone and impurities in clay has a positive influence.

Good flowable concrete can be obtained with use of superplasticizers.

In some formulation, SP dosage may even be less.

No segregation, no bleeding.

Further improvements are possible with PSD optimisation, grinding aids, etc.
Carbonation

Indoor

3D
PC
PPC30
LC\textsuperscript{3}-50

28D

Outdoor

3D
PC
PPC30
LC\textsuperscript{3}-50

28D

2 years natural conditions: similar to other blends
Carbonation

Reducing calcium content; reduces buffer to carbonation

\[
\text{CaO} + \text{H}_2\text{O} \rightarrow \text{Ca(OH)}_2
\]

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

All CaO content can react with CO\(_2\), not just portlandite

What about colour
» Intensity determined by iron concrete

» Red to grey by kiln atmosphere
Colour control. IPIAC technology

Calcination with reduction during cooling
Concluding remarks

➢ Future cements will be based on Portland cement clinker with increasing levels of incorporation of SCMs

➢ Calcined clays are the only realistic option for extending the use SCMs

➢ Possible to obtain similar mechanical properties to OPC / CEM I with 50% clinker and clays with >40% kaolinite

➢ Calcined clays have very positive impact on:
  ▪ Chloride ingress
  ▪ ASR

➢ If we are serious about more sustainable concrete we need to use cements with lower CO₂ emissions, e.g LC³ clinker/ calcined clay / limestone blends

➢ Europe has an important role to play in facilitating uptake worldwide: standards and research
Thank you

More information on: www.LC3.ch

Sign up for the LC3-newsletter and follow us on:

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LC3-Low Carbon Cement
LC3-Limestone Calcined Clay Cement

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