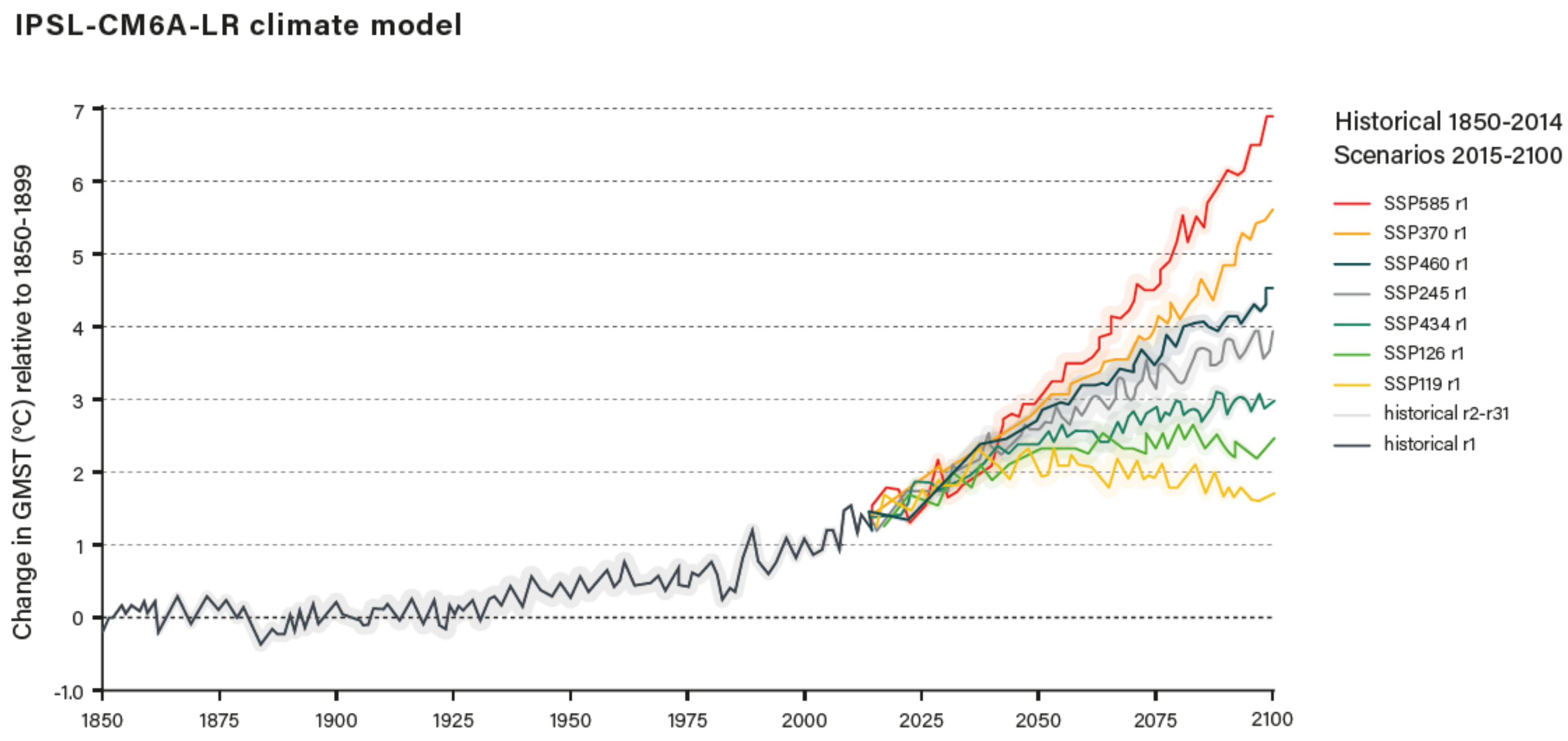
LC3 / LC2:

major CO_2 mitigation levers for the cement industry

Speed & Scale

Professor Karen Scrivener, FREng, EPFL, Switzerland

We need to act fast



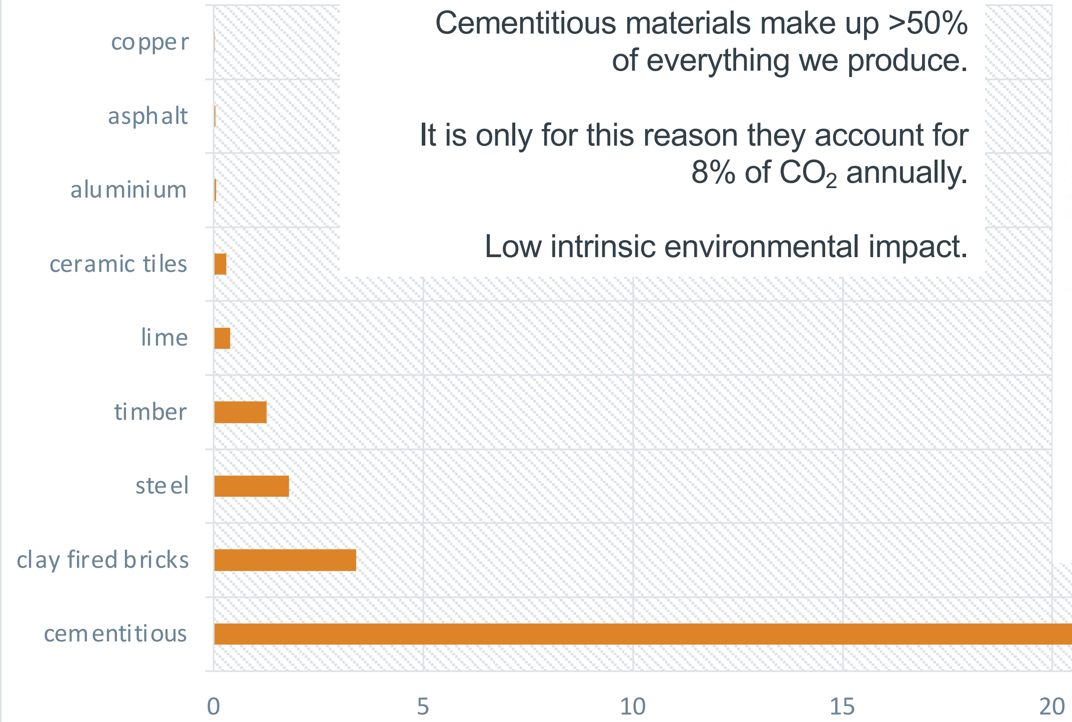
Source: Jean-Marc JANCOVICI, jancovici.com

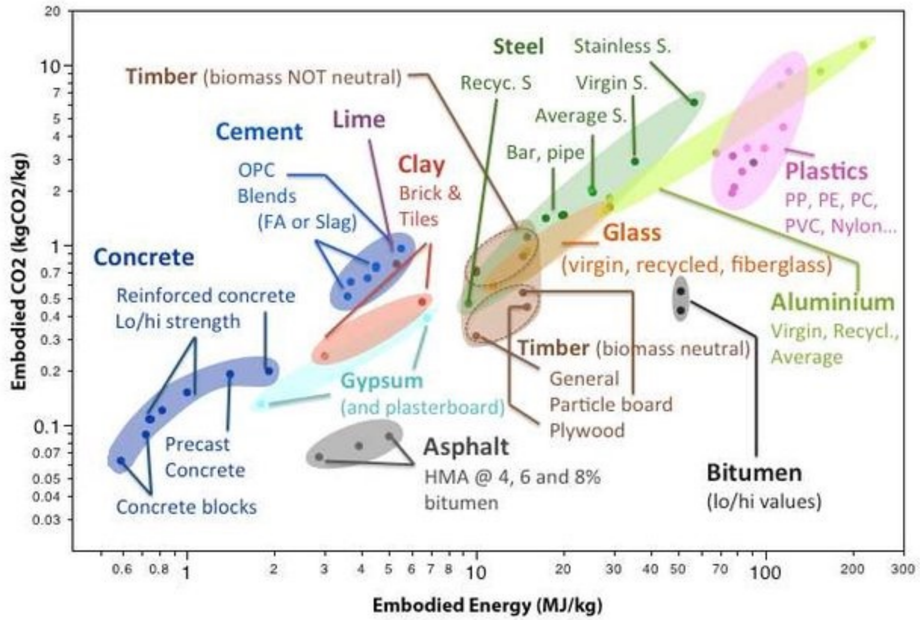
Concrete + Mortar are irreplaceable

Cementitious materials make up >50% of everything we produce.

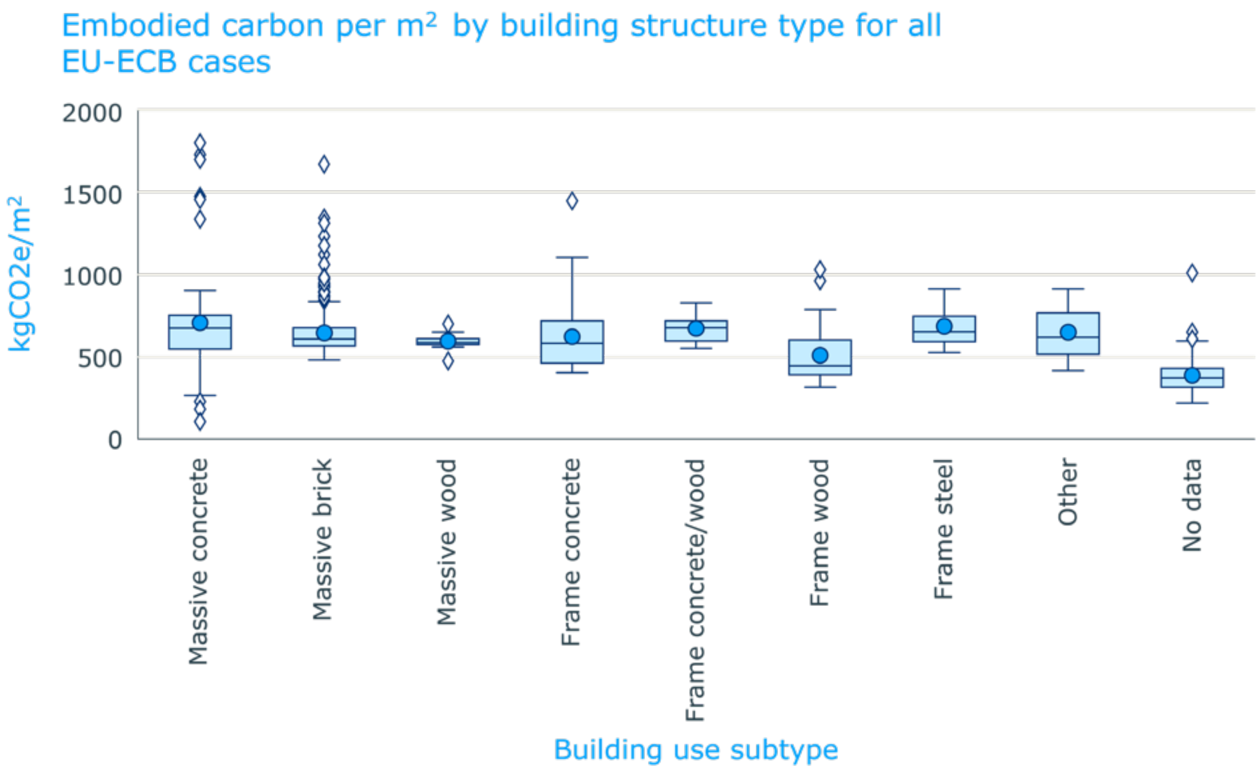
It is only for this reason they account for 8% of CO_2 annually.

Low intrinsic environmental impact.





Would it help to replace concrete by other materials?

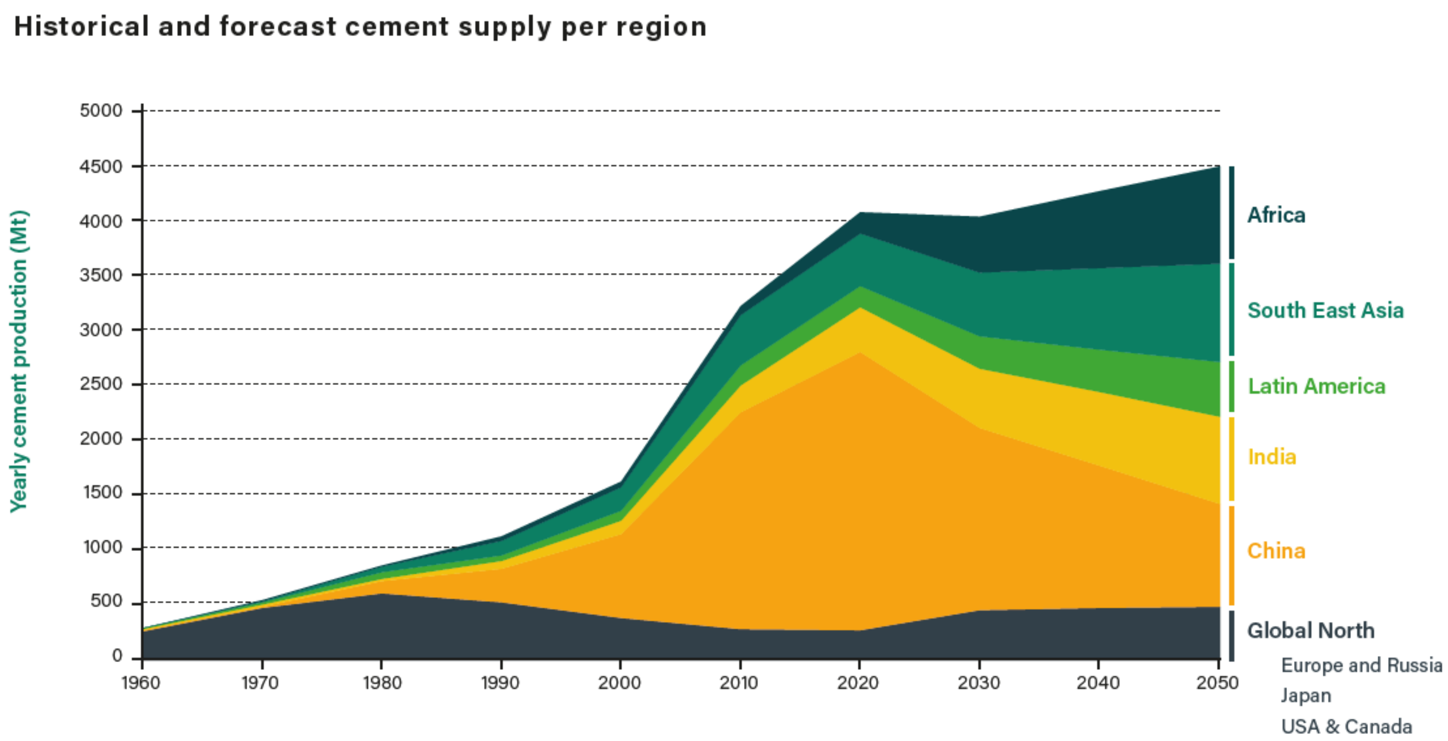


Röck M, Sørensen A, Tozan B, Steinmann J, Le Den X, Horup L H, Birgisdottir H
Towards EU embodied carbon benchmarks for buildings – Setting the baseline: A bottom-up approach, 2022, https://doi.org/10.5281/zenodo.5895051.

Limestone Calcined Clay Cement LC3

LOW CARBON LOW COST LOW CAPITAL

Demand for cement in the Global South



We need solutions for people in developing countries

EPFL

Limestone Calcined Clay Cement LC3

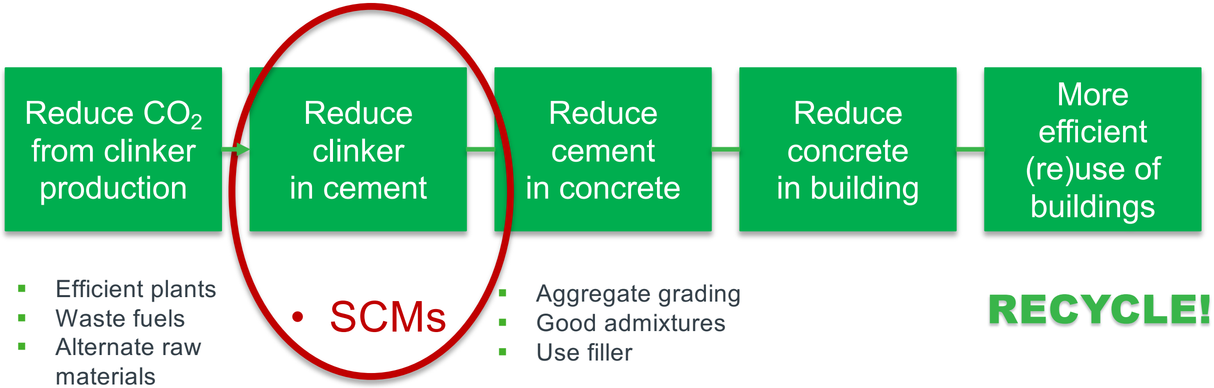
LOW CARBON LOW COST LOW CAPITAL

Report for European Climate Foundation 2017

A SUSTAINABLE FUTURE FOR THE EUROPEAN CEMENT AND CONCRETE INDUSTRY

Technology assessment for full decarbonisation of the industry by 2050

ETH zürich



Substantial reductions in emissions > 80% can be achieved by working through the whole value chain.

If only cement level is considered not more than about 50% possible without carbon capture and storage.

EPFL

nature communications

Article https://doi.org/10.1038/s41467-023-40302-0

Near-term pathways for decarbonizing global concrete production

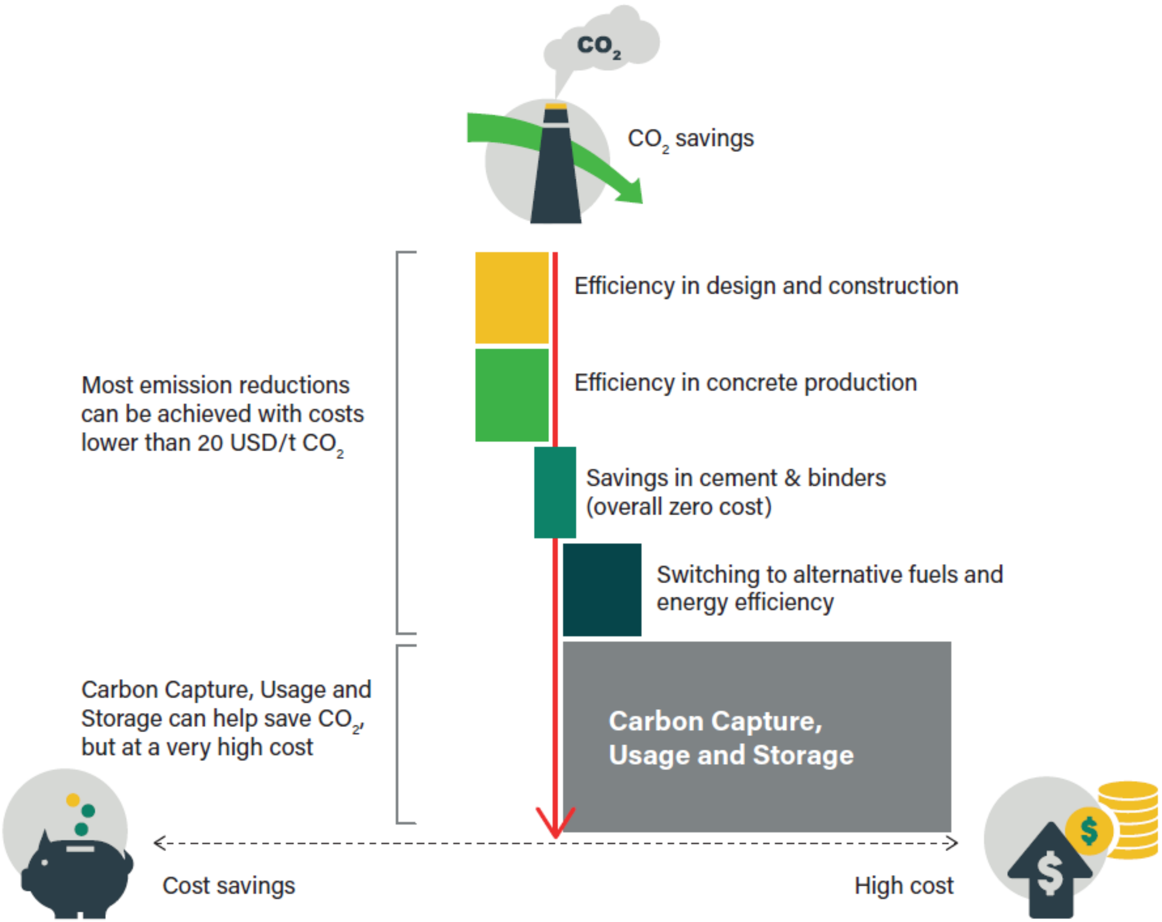
Received: 27 January 2023

Accepted: 21 July 2023

Josefine A. Olsson[1], Sabbie A. Miller[1] ✉ & Mark G. Alexander[2]

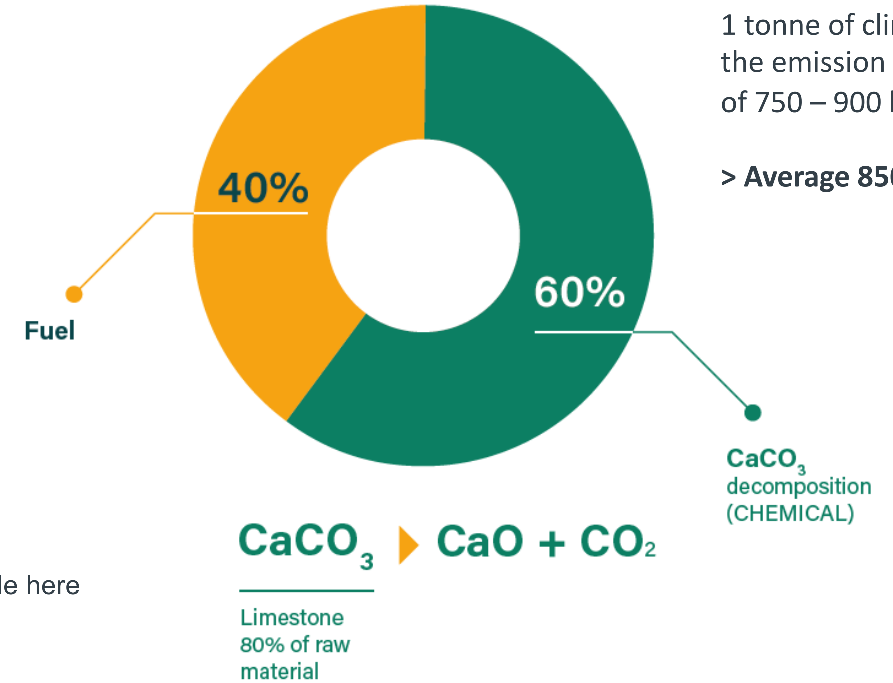
Calculated 76% with these strategies

Getting to net zero can be done with little to no cost



Origins of CO_2 emissions in clinker production

- The production process is highly optimised up to around 80% of thermodynamic limit.
- It is estimated that **< 2%** further savings can be made here
- Use of waste fuels, which can be **> 80%** reduces the demand for fossil fuels



1 tonne of clinker leads to the emission of 750 – 900 kg CO_2

> Average 850kg/t

No silver bullet

Despite the media interest they attract, most niche technologies – such as alkali activated materials, cement from algae, etc are:

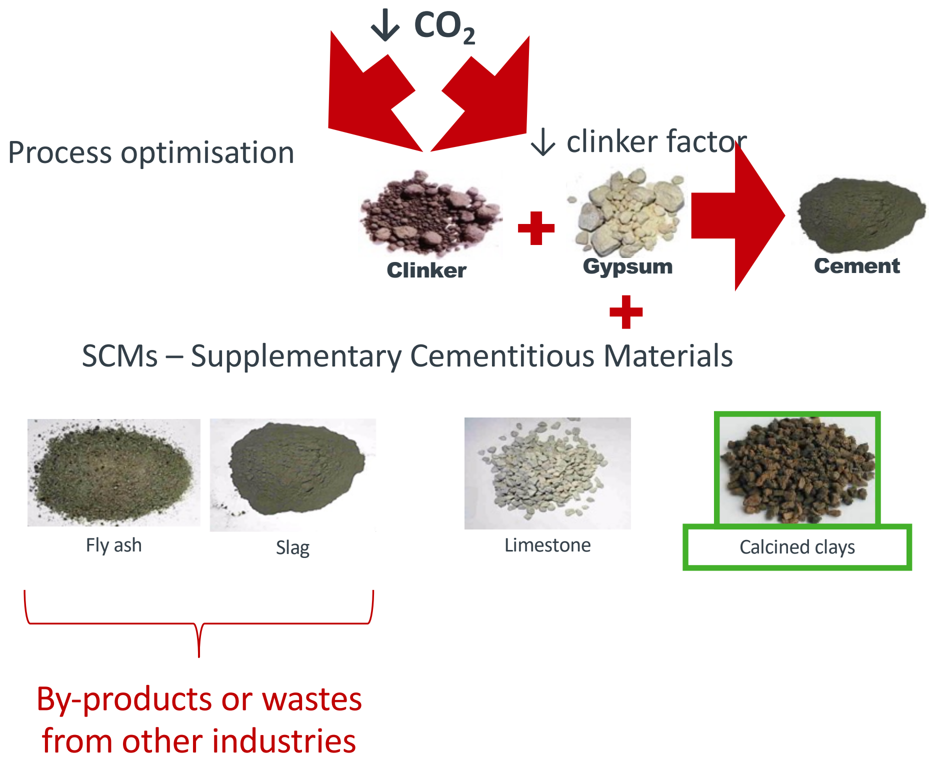
- impractical,
- costly,
- unscalable,
- will take too long to mature

so have little to no possibility of delivering any significant impact.

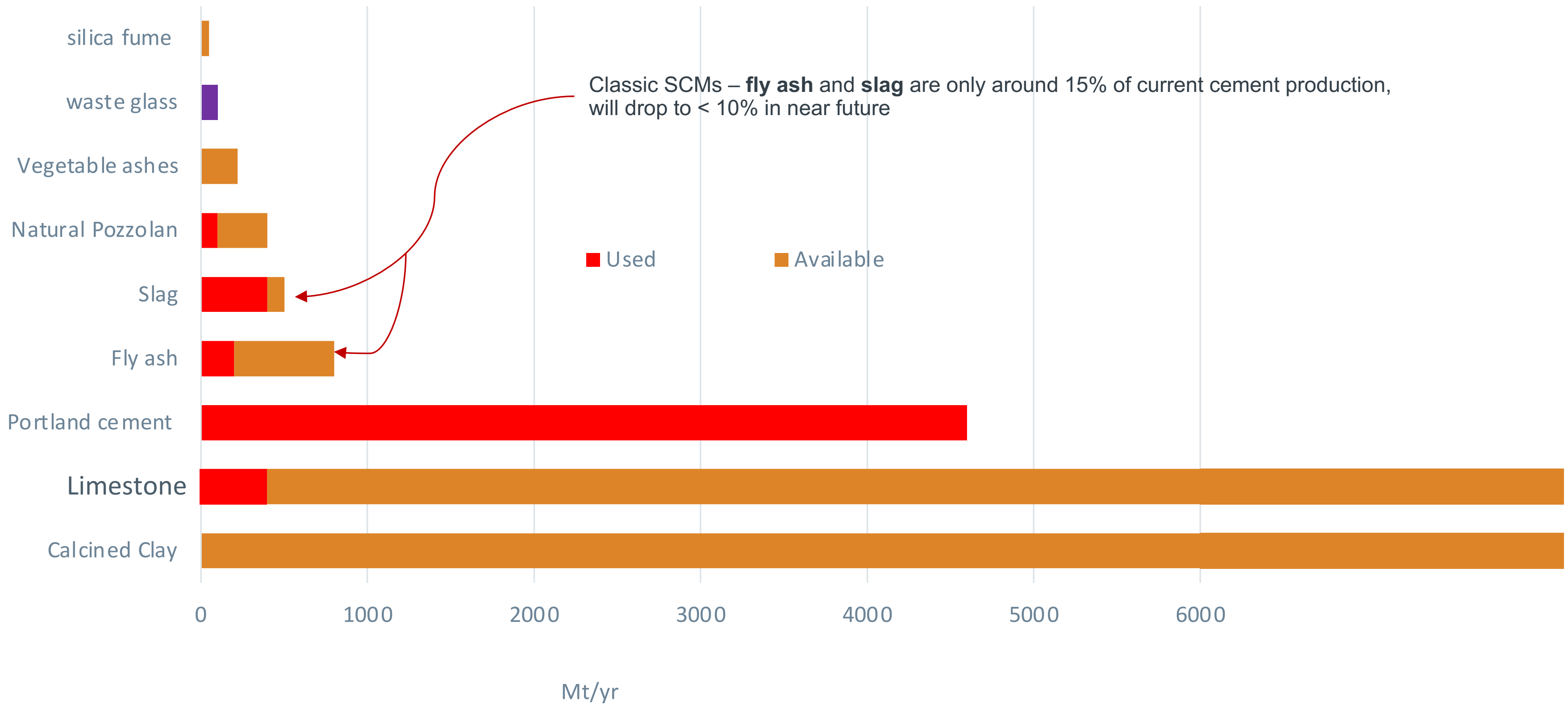
Portland based cements
will continue to dominate.

Blended cements are the most
realistic option to reduce CO_2
and extend resources.

Most promising approach – reducing the clinker factor



Availability of SCMs



There is no magic solution

- Blended with SCMs will be best solution for sustainable cements for the foreseeable future.
- **Only material** really potentially available in viable quantities **is clay**.
- **Synergetic reaction** of calcined clay and limestone allows high levels of substitution
- EPFL led the LC^3 Project supported by **Swiss Agency for Development and Cooperation (SDC)**, 2013-2022.
- **Climateworks Foundation** supporting the LC^3 Project since 2022.

LC3 PROJECT

Schweizerische Eidgenossenschaft
Confédération suisse
Confederazione Svizzera
Confederaziun svizra

Swiss Agency for Development and Cooperation SDC

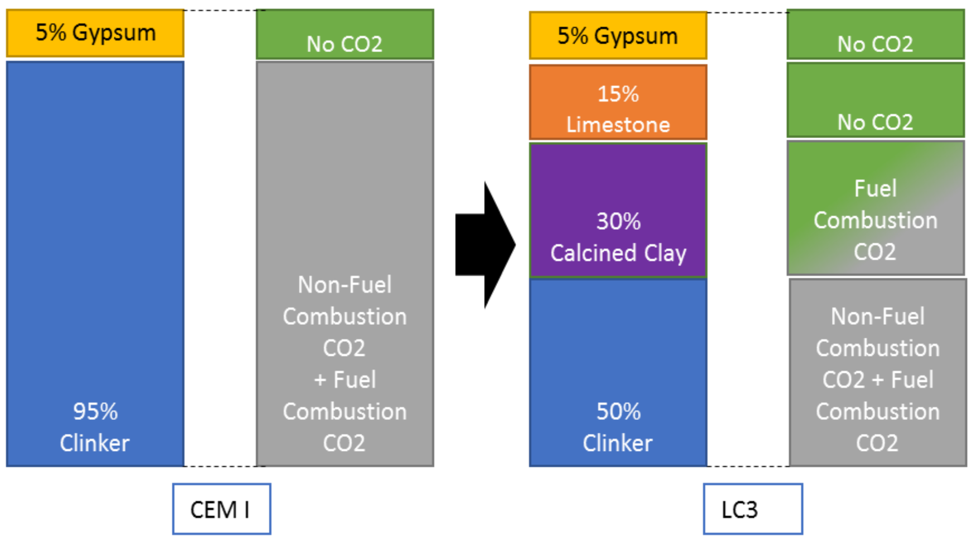
climateworks FOUNDATION

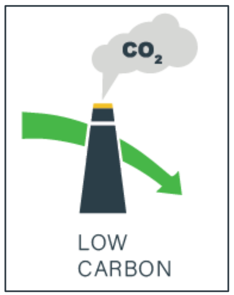
EPFL

What is LC^3 ?

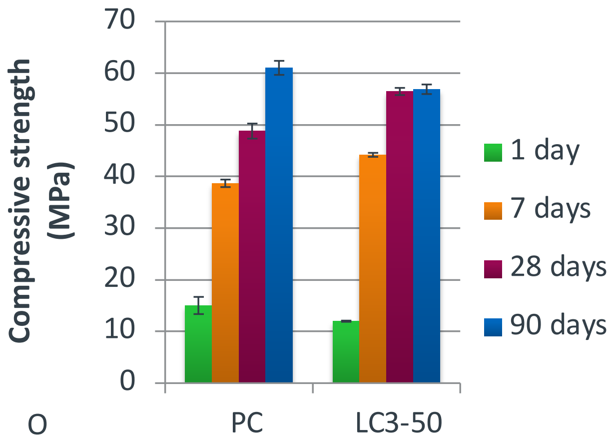
Landscape of Partners LC3 PROJECT

How does LC^3 reduce emissions?





LC^3 has comparable strength to OPC

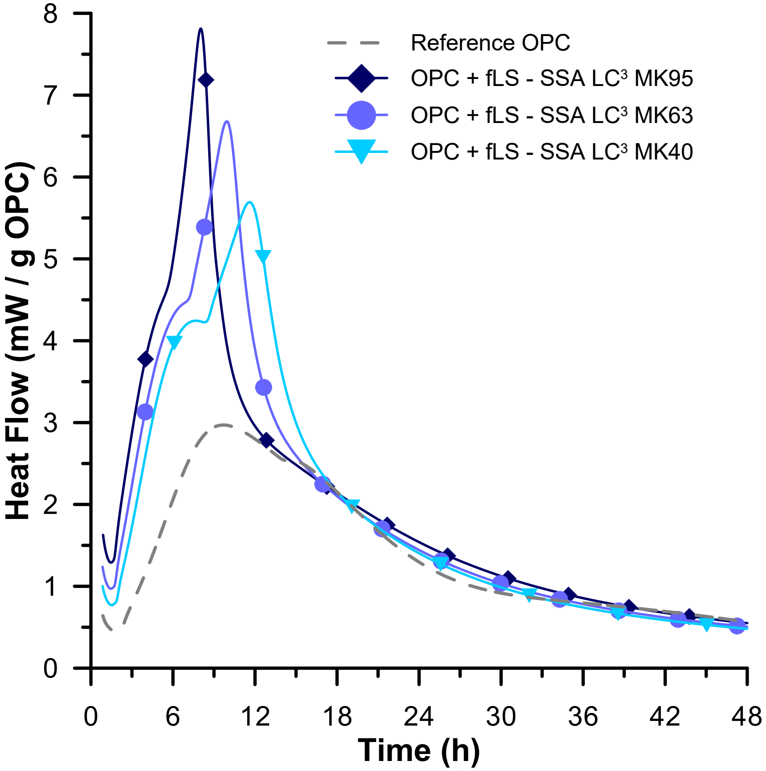


LC3-50 = 50% clinker.

- 50% less clinker
- 40% less CO_2
- Similar strength
- Better chloride resistance
- Resistant to alkali silica reaction

Sulfate adjustment in blended cements

OPC with different amounts of fine limestone

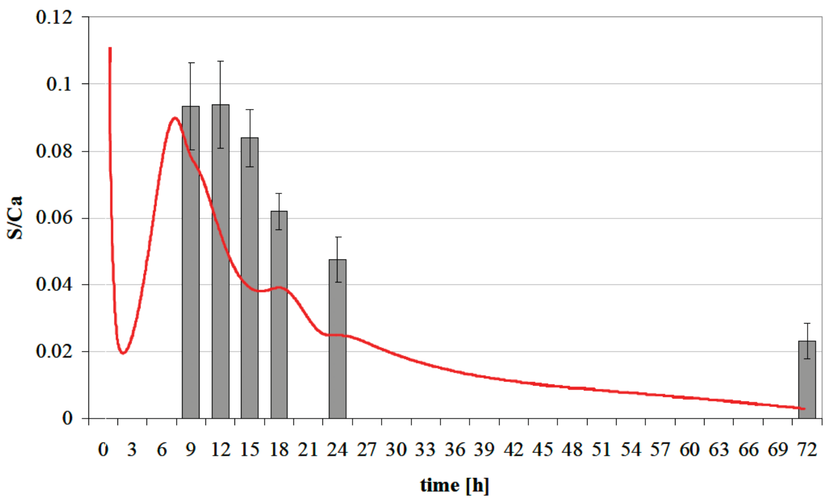


- Experiment aimed to replicate the specific surface area of LC^3, without the addition of any aluminate phases
- As observed, the effects on the aluminate reaction are comparable or even more pronounce than the calcined clay addition
- This means that the time of occurrence of the aluminate reaction, linked to gypsum depletion, is overwhelmingly controlled by the binder's **specific surface area**

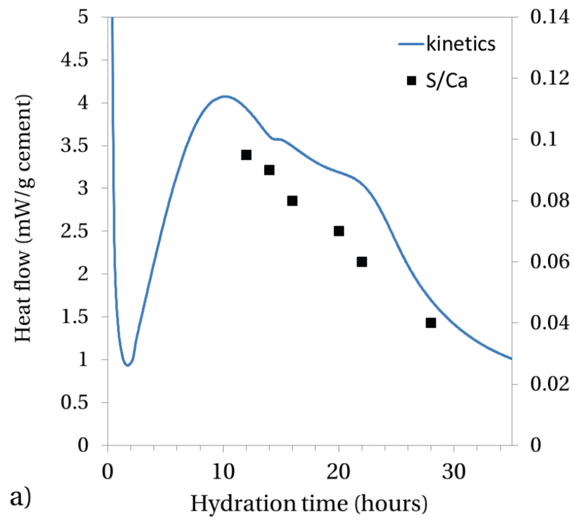
Filler effect : accelerated rate of reaction of C_3S, leading to an increased amount of C-S-H formed

The interaction of sulfate with C-S-H

- C-S-H is the main reaction product of the hydration of cement
- It is well know that it can adsorb/incorporate different ions on its surface/structure (Al, Na, K, Cl, S)



Quennoz, PhD Thesis, 2011



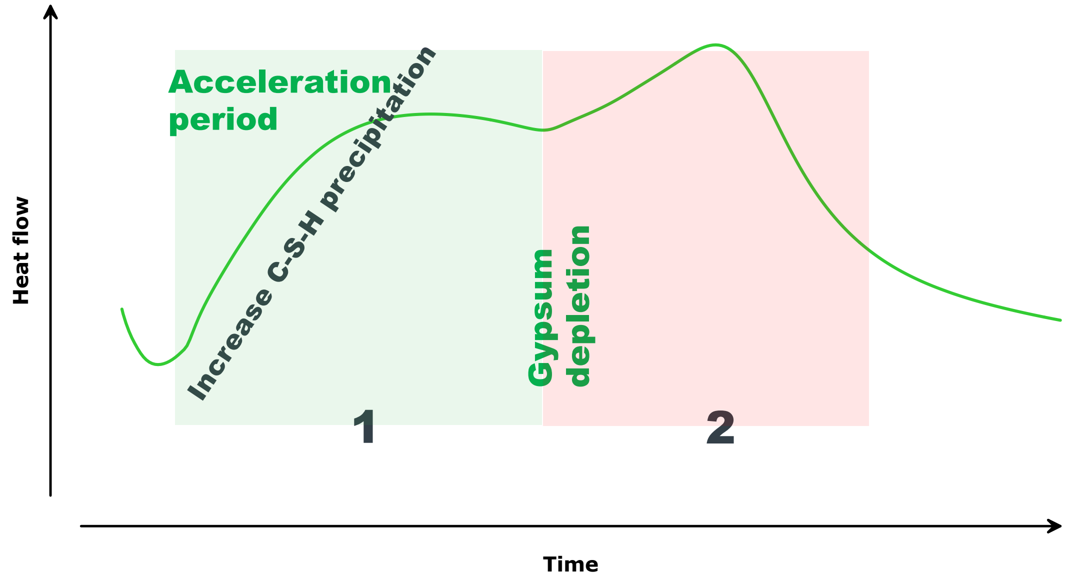
Berodier, PhD Thesis, 2015

Limestone Calcined Clay Cement LC3

LOW CARBON LOW COST LOW CAPITAL

Sulfate adjustment in blended cements

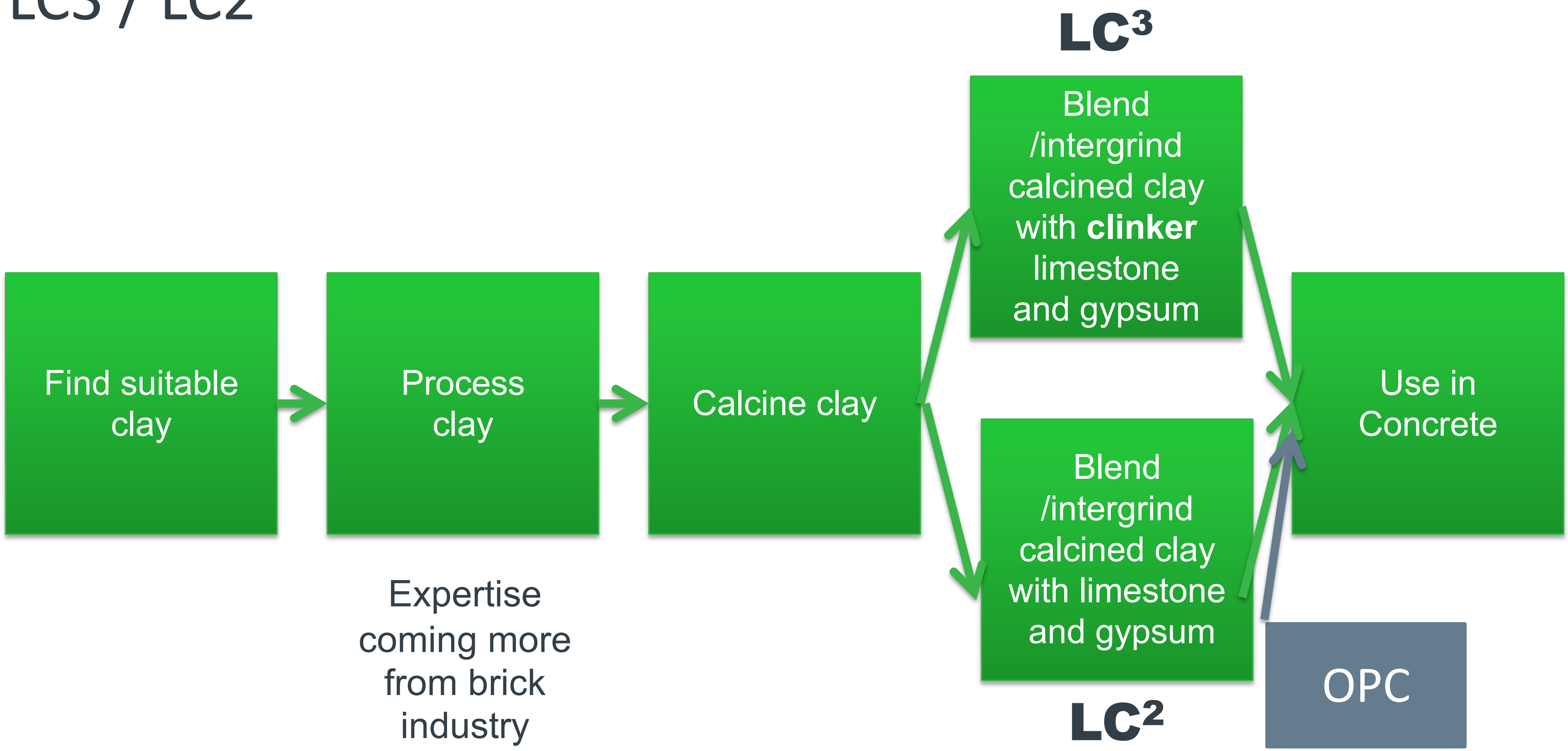
Proposed Mechanism



1. Sulfate gets adsorbed in the C-S-H surface. If the rate of C-S-H precipitation is increased, the rate of sulfate adsorption also increase
   i. Finer material hydrate quicker, consuming more sulfate per unit time
   ii. The addition of more nucleation surface (fillers) affect the C-S-H precipitation rate and therefore the sulfate balance, but the effect is independent of the chemical composition of the filler (in particular, of the Al content)
2. As hydration keeps ongoing, gypsum is depleted, triggering the desorption of sulfate from C-S-H which then reacts with aluminates to form ettringite (second ettringite formation)
   i. The extent and rate of the reaction is a function of how much sulfate gets desorbed from C-S-H (and maybe the space filled already at this time)

LOW CARBON
LOW COST
LOW CAPITAL

LC3 / LC2



Expertise coming more from brick industry

Limestone Calcined Clay Cement LC3

LOW CARBON LOW COST LOW CAPITAL

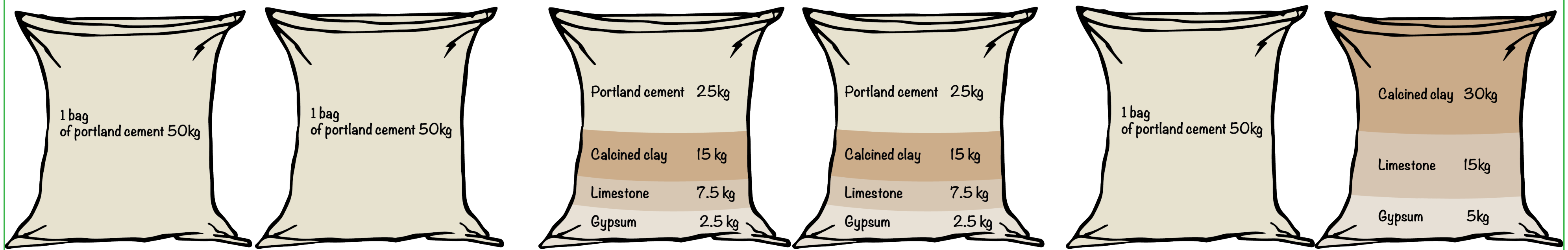
LC3
Blend at cement plant

LC2
Blend at ready mix plant

<u>Ordinary Portland Cement : 100 kg = 2 bags</u>
Conventional solution

<u>LC3: 100 kg = 2 bags</u>
(pre-mixed at factory) Needs new standards in most countries
Clinker factor = 50 %

<u>LC2: 100 kg = 1 bag Portland + 1 bag LC2</u>
(mixed at concrete ready mix site) Already possible with existing standards in most countries, Clinker Factor also 50 %



CO_2 emissions = 80 kg per 100 kg

CO_2 emissions = 50 kg per 100 kg

CO_2 emissions = 50 kg per 100 kg

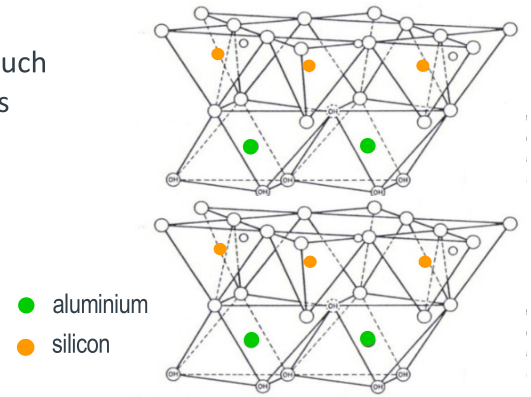
EPFL LMC

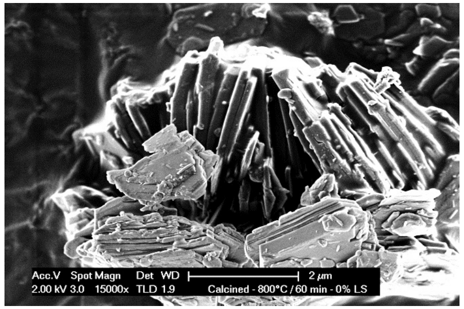
Limestone Calcined Clay Cement LC3

LOW CARBON LOW COST LOW CAPITAL

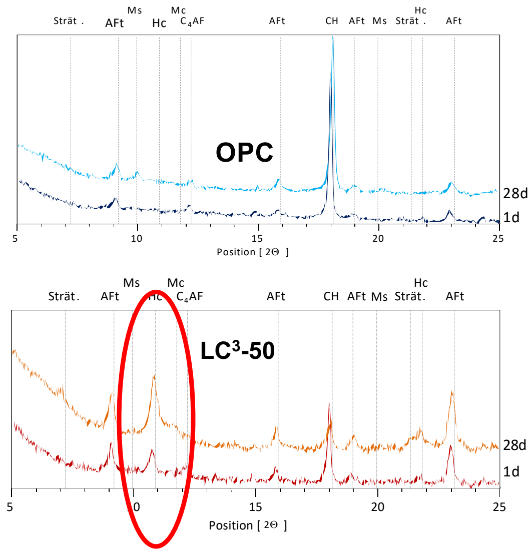
Why can we get such high replacement levels?

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





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



HIGH PERFORMANCE

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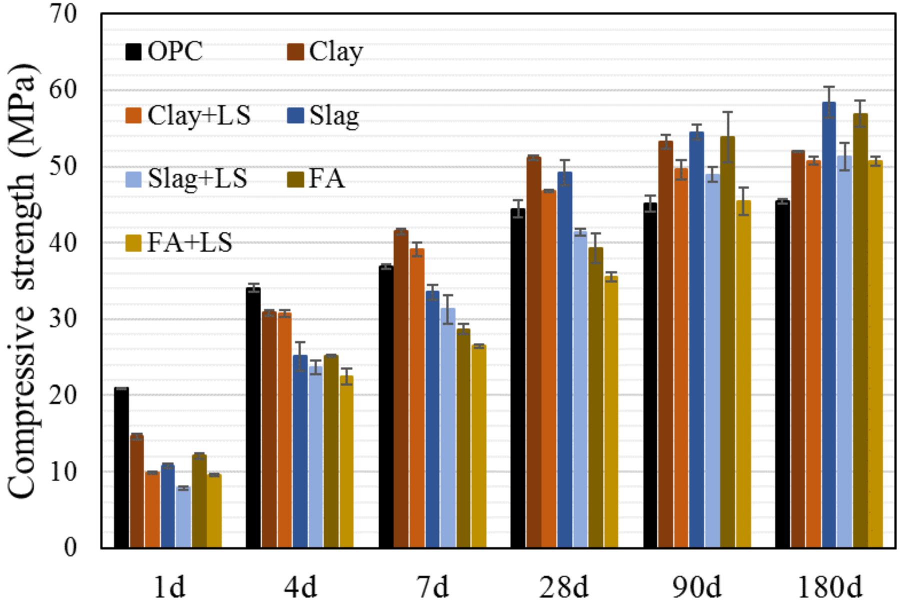
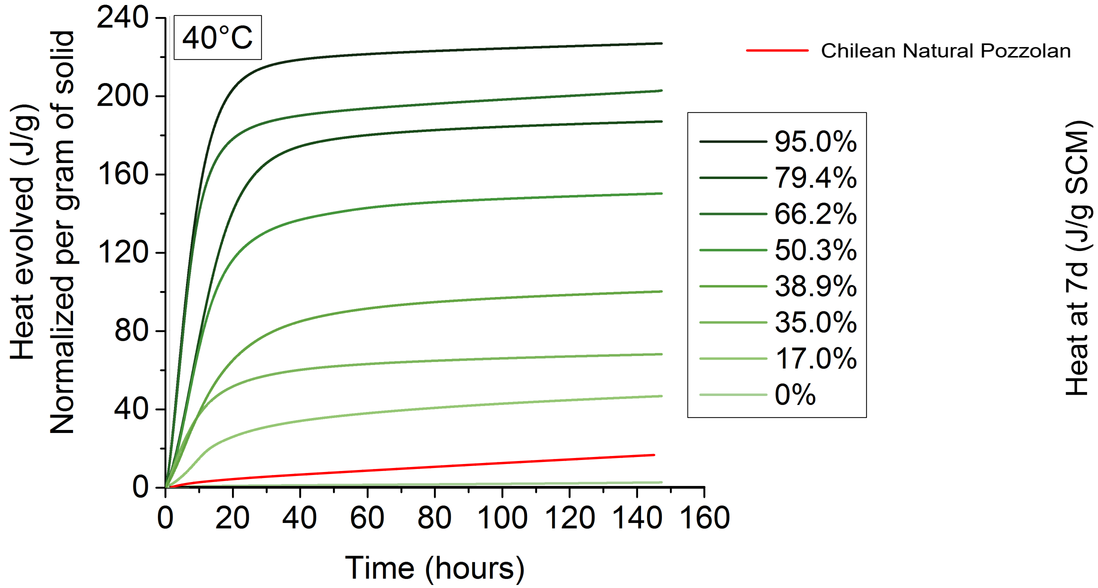
Comparison of calcined kaolinitic clay, slag and fly ash



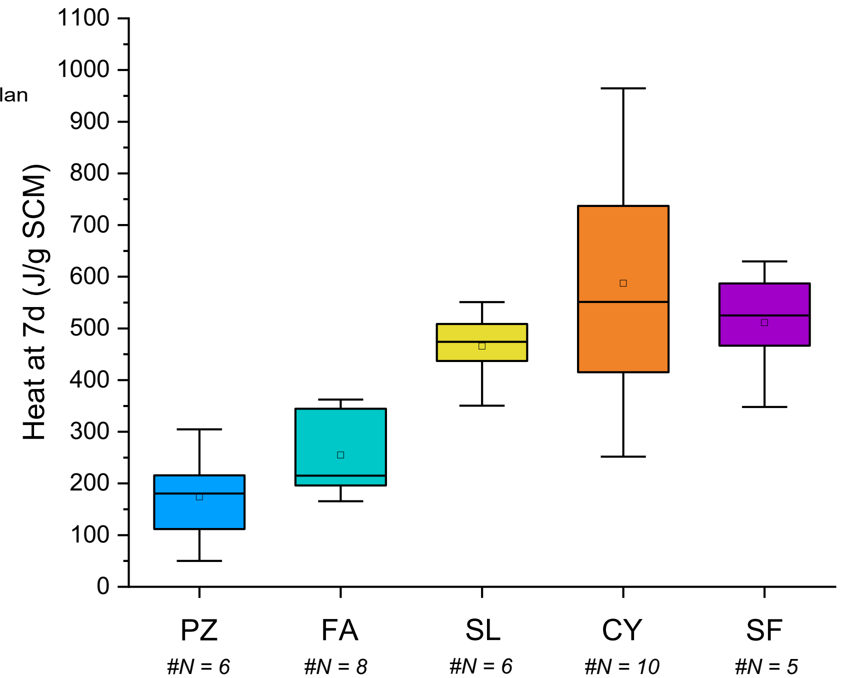
Binary systems 70% clinker, 30% SCM
Ternary systems, with limestone 50% clinker, 30% SCM, 15% limestone

Calcined clay vs. other pozzolans

natural pozzolan (R^3 test)



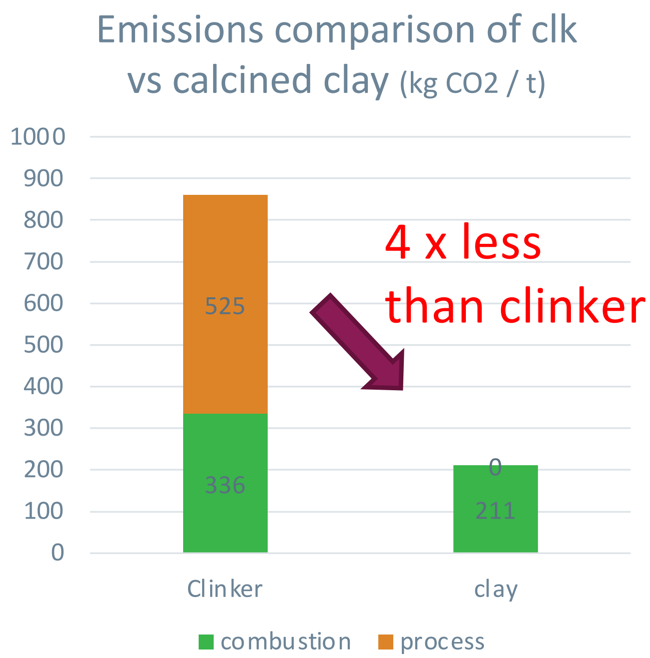
ASTM C1897



Kaolinitic clay with the lowest kaolinite content is more reactive than most fly ashes commonly used in the industry!!

F. Zunino, RILEM TC-267 TRM

CO_2 emission savings clinker vs. clay



Emissions from clinker

Emissions from combustible

- ✓ Thermal consumption from clinker: 3.5 GJ /T clinker (GNR world 93AG)
- ✓ CO2 emissions from coal: 0.096 T CO2 / GJ (WBCSD / CSI value)

= CO2 emission from combustible: 336 kg CO2 / T clinker

Emissions from process

= CO2 emission from process: 525 kg CO2 / T clinker (IPCC default value)

Total emissions clinker: 861 kg CO2 / T

Emissions from calcined clay

Emissions from combustible

- ✓ Thermal consumption from clay: 2.2 GJ /T clay (real case)
- ✓ CO2 emissions from coal: 0.096 T CO2 / GJ (WBCSD / CSI value)

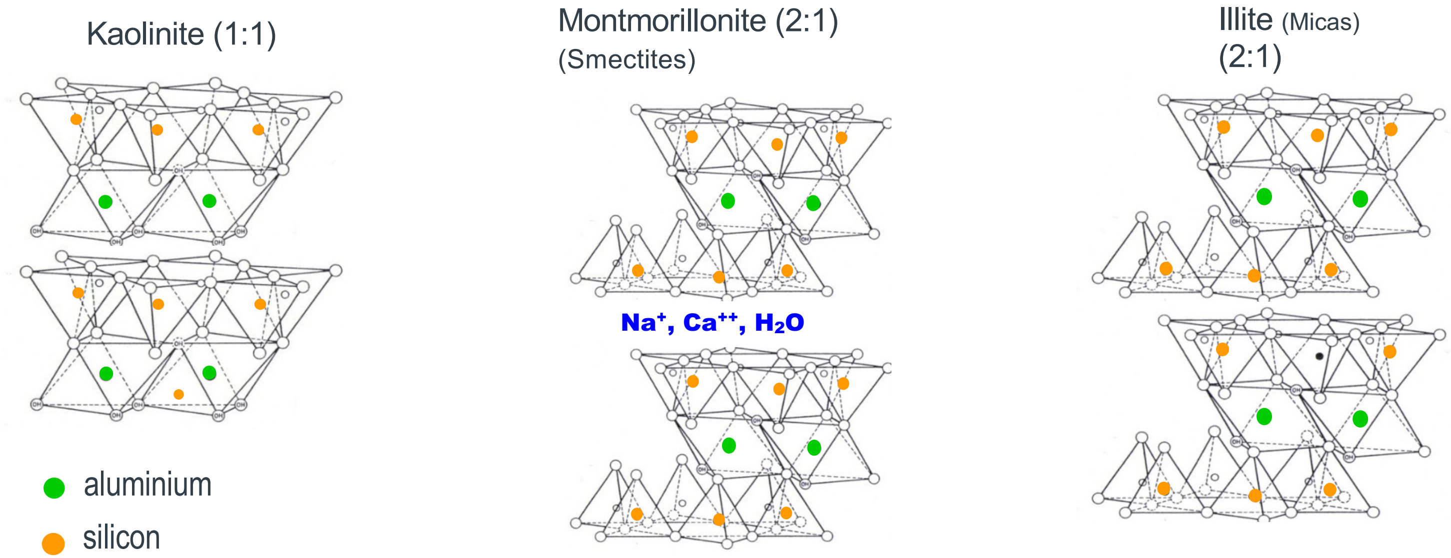
= CO2 emission from combustible: 211 kg CO2 / T clay (-37% vs. clinker)

Emissions from process: **0**

Total emissions clay: 211 kg CO2 / T (4 x time less than clinker)

What kinds of clay are suitable?

Three basic clay structures

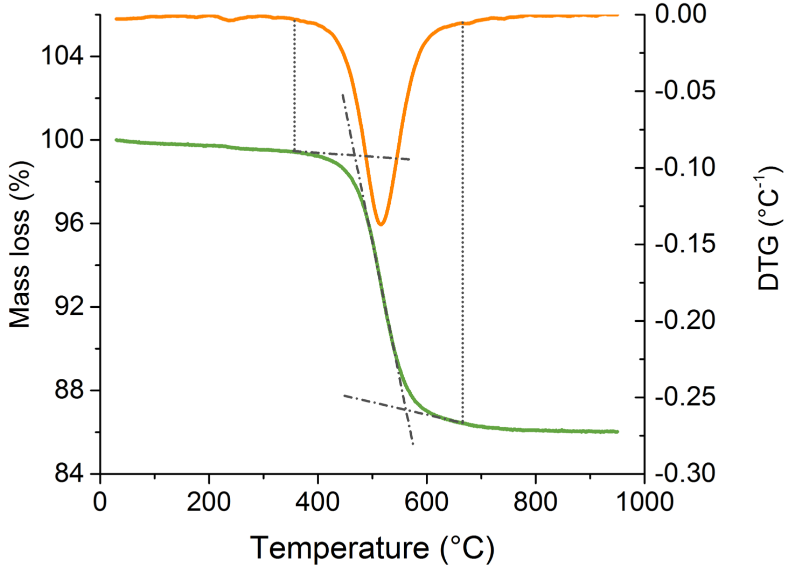


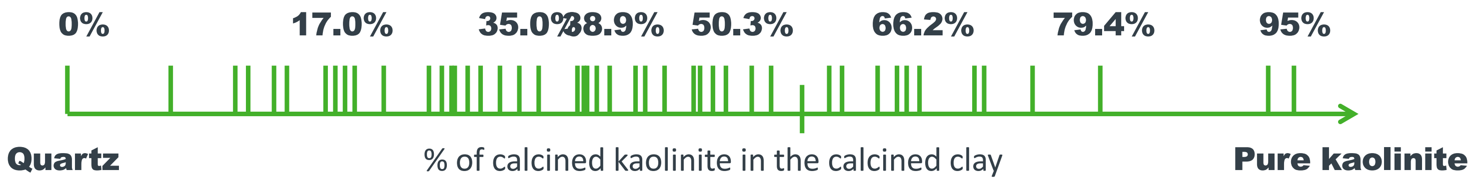
“Metakaolin”, sold as high purity product for paper, ceramic, refractory industries
Requirements for purity, colour, etc, mean expensive 3-4x price cement

Clays containing metakaolin available as wastes
– over or under burden NOT agricultural soil
Much much less expensive often available close to cement plants

Over 100 clays studied from around the world

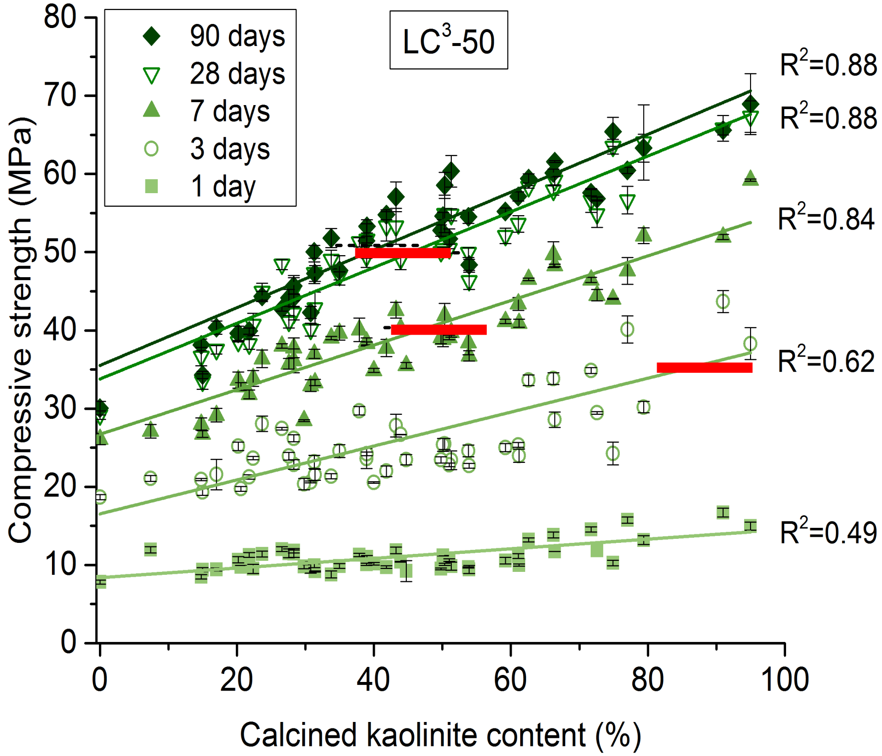
Different calcination conditions
Different compositions, impurities
Different physical properties





Benchmark test of clay strength

- Compressive strength EN 196-1 at 1, 3, 7, 28, 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 30-60%

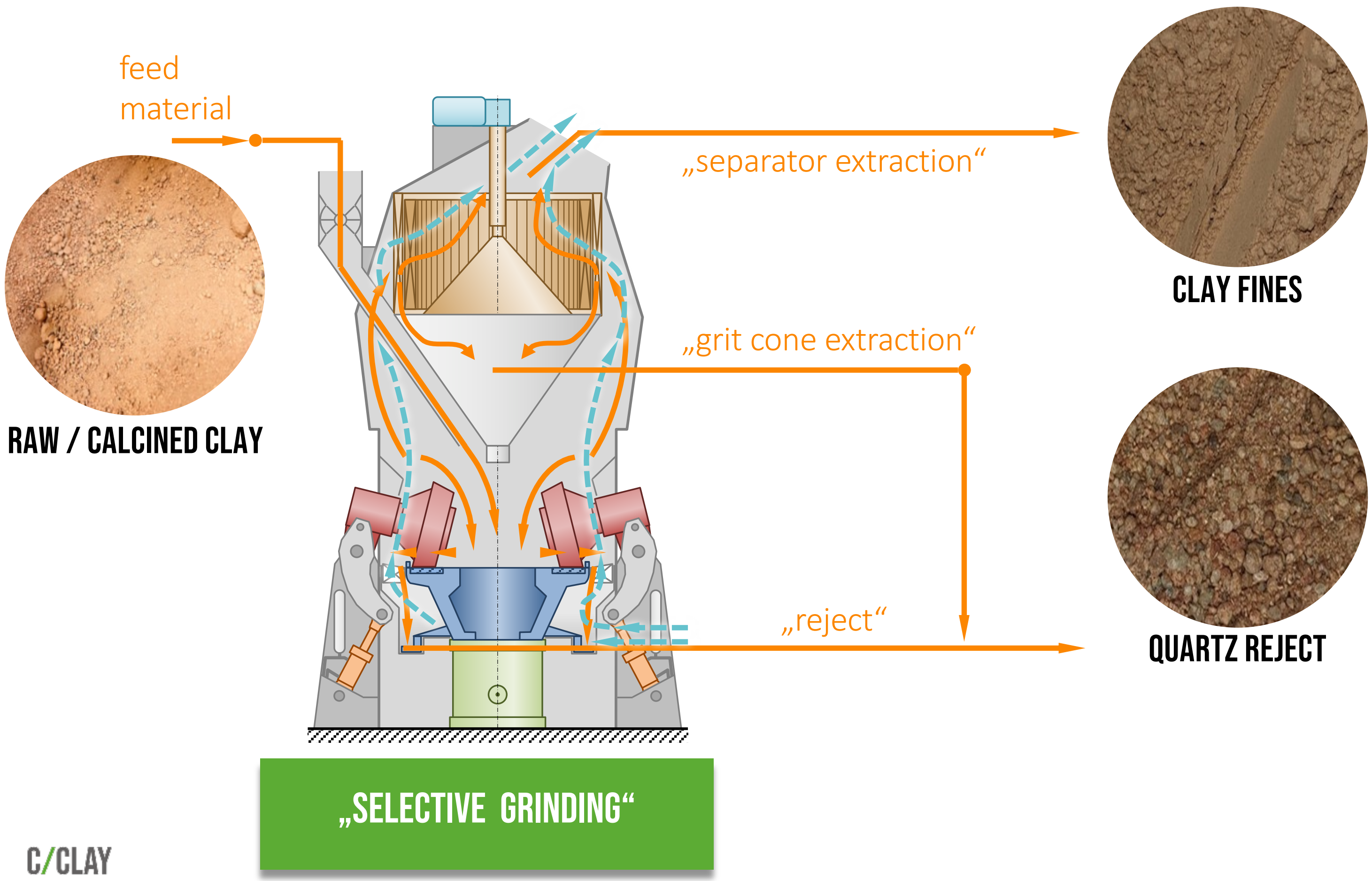
- Higher contents, possible to use more limestone
- *Even better economics and ecology*

- Lower contents can be enriched by separation
- *Separated, fine quartz can be sold as separate product*

- These kind of materials are not documented in official reserves of “kaolin”
- The amounts of suitable materials 100s of times higher than such reserves

Grinding clay

Quartz separation via selective grinding

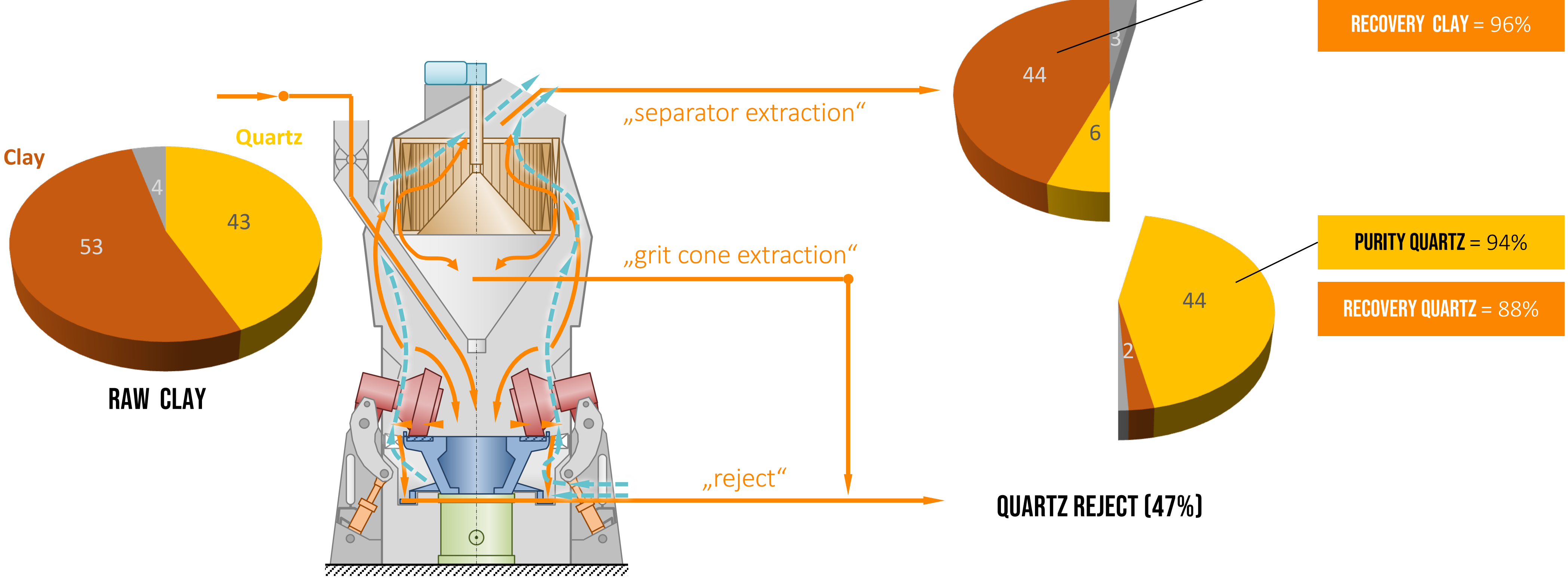


ADVANTAGES

- clay enrichment
- utilization of mid-quality clays
- better burnability
- better heat transition
- less energy consumption (thus CO_2 and OPEX savings)
- better reactivity index
- better cement/concrete performance

Grinding clay

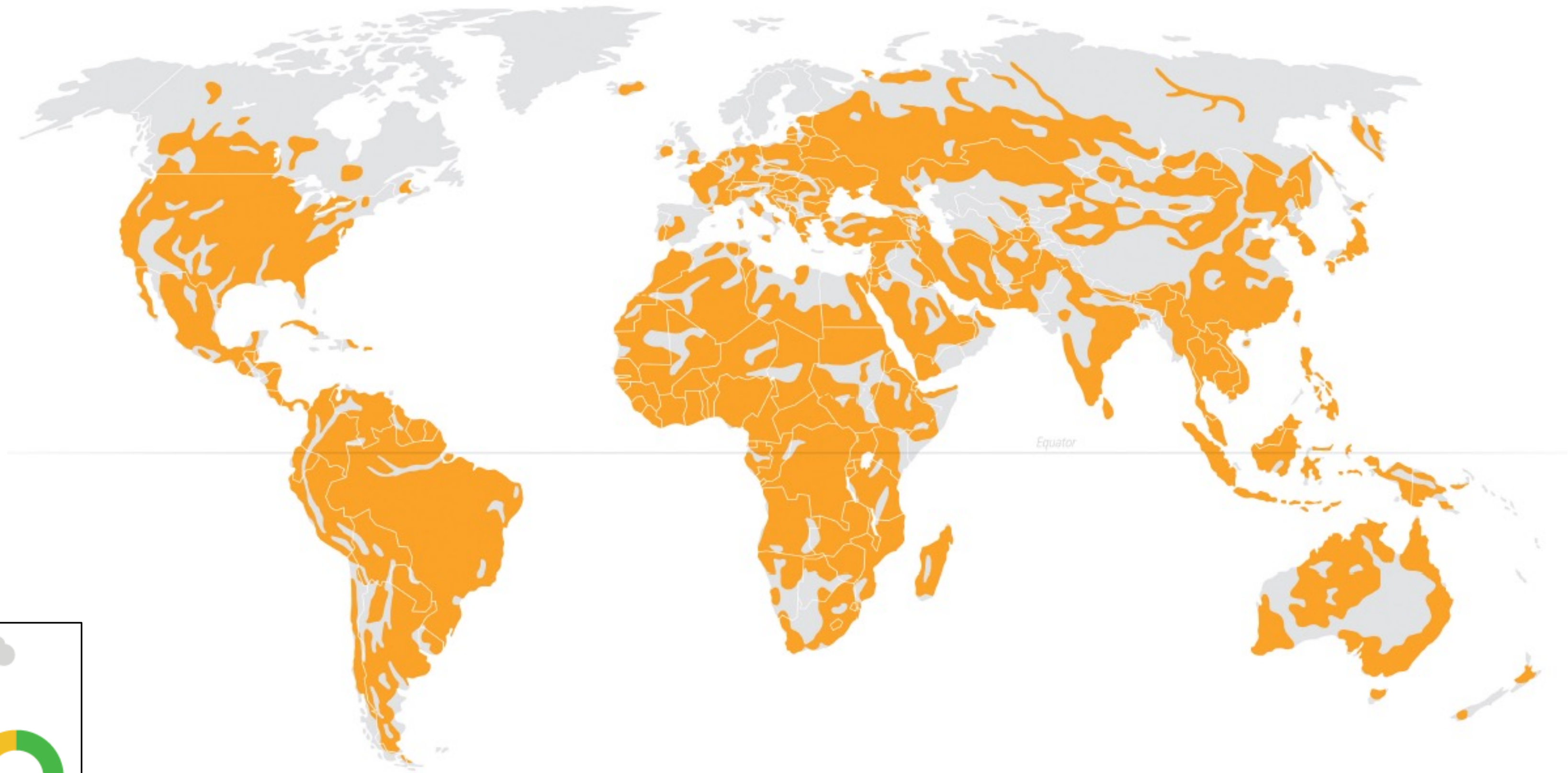
Quartz separation via selective grinding



LOESCHE
INNOVATIVE ENGINEERING

World distribution of kaolinitic clays

LOW CARBON
LOW CAPITAL
HIGH PERFORMANCE
SCALABLE



Source: Ito and Wagai, Scientific data 2017

LC3 INNOV

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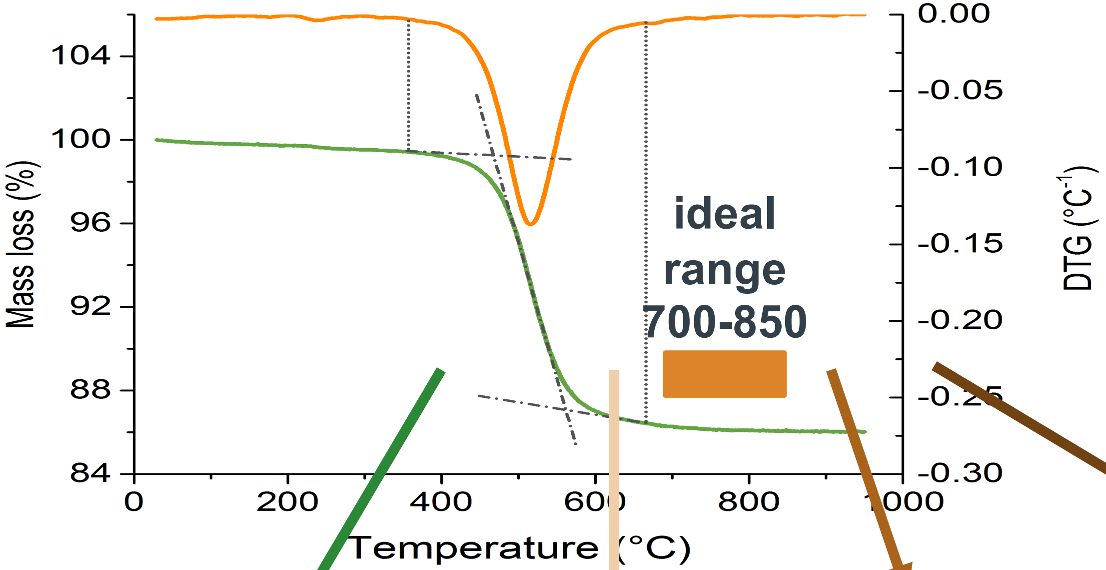
Calcination of clay

Temperature window 700-850°



<600
Not reactive

600-700
dehydroxlation complete but reactivity increases

850-1000
Surface area reduced reactivity decreases

>1000
crystalline phases formed
no reactivity

Calcination methods:

Rotary kiln

- Advantages
  - Robust
  - Tolerant to moisture content up to 20%
  - Fairly large particles (few mm) can be calcined efficiently
  - Easy colour control technology

- Disadvantages
  - Reputed to have higher energy consumption

But real kiln built in Ivory Coast has energy of 550 kCal/ kg, 2.3 MJ/kg

Flash calcination

- Advantages
  - Calcination more energy efficient
  - Lower opex?

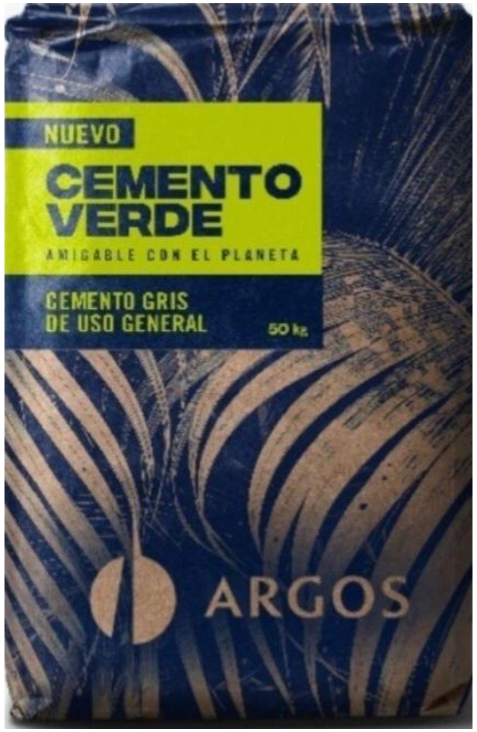
- Disadvantages
  - Very low moisture (<5%)
    - Dryer and dry storage silo required
  - Small particles
    - crusher

To date testing many clays indicates no significant difference in reactivity

Limestone Calcined Clay Cement LC3

LOW CARBON LOW COST LOW CAPITAL

Industrial projects: Cemento Verde ARGOS, Colombia



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Industrial projects: CIMPOR, Ivory Coast

Colour control at Ivory Coast plant

Demonstration structure

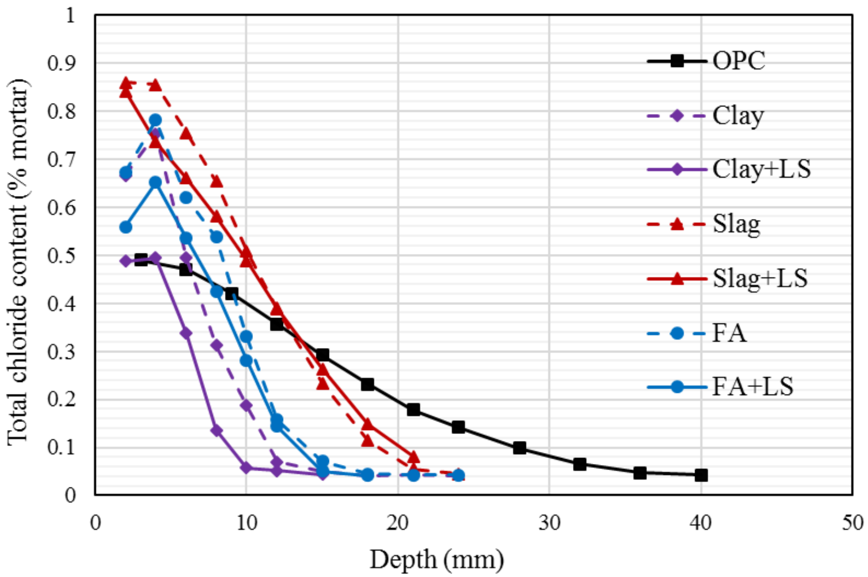
Around 14 tonnes of CO_2 saved
Compared to existing solutions

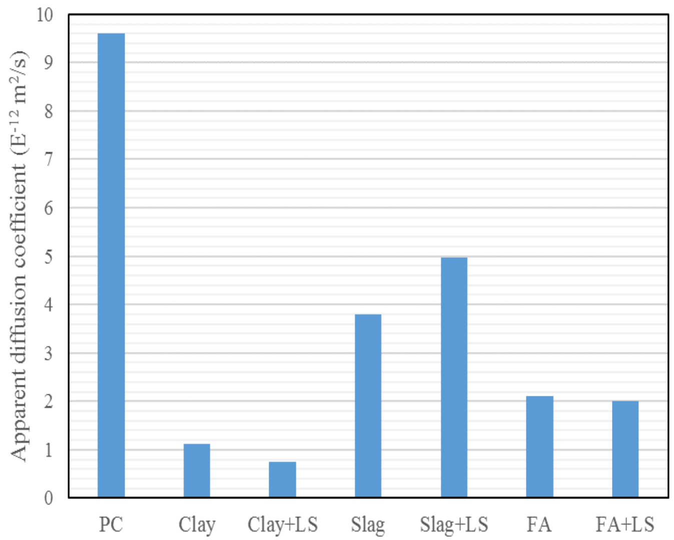
Argos Colombia

Key Advantages

- Chloride resistance
- Suppression of alkali silica reaction

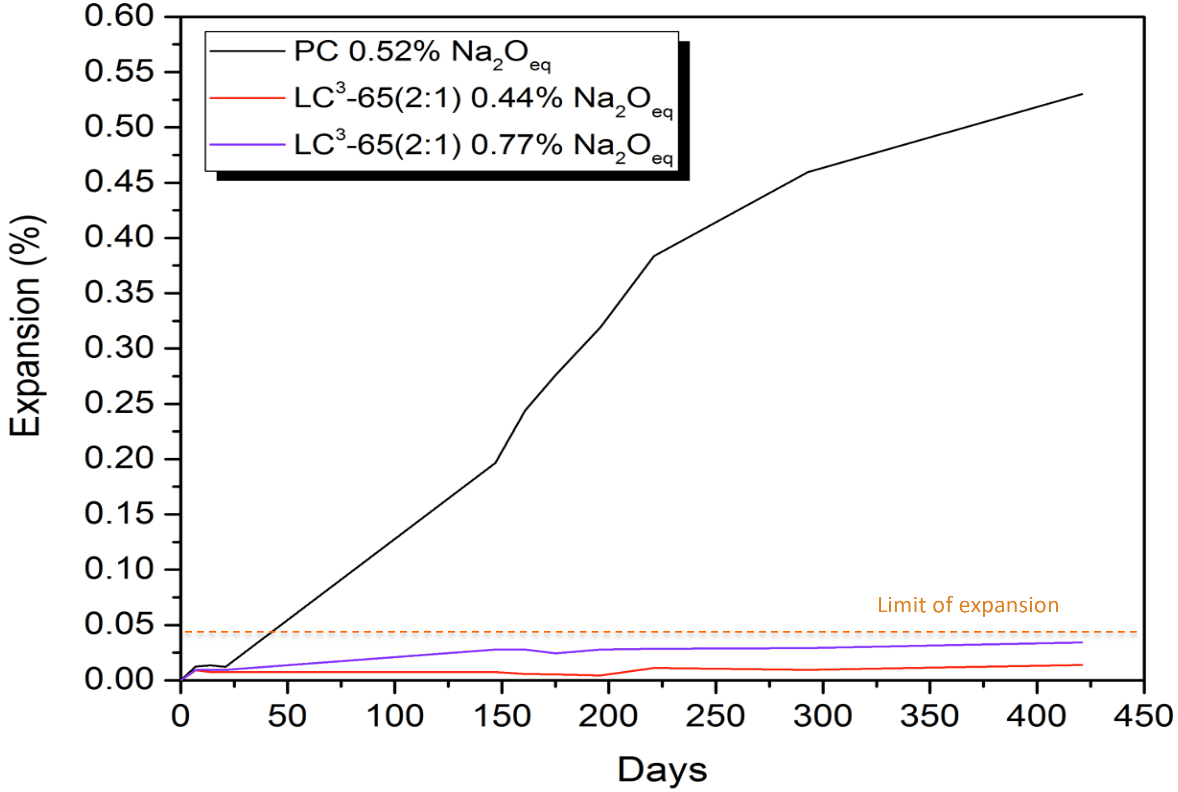
Chloride ponding ASTM





Apparent diffusion coeffs.

Alkali silica reaction



Perceived problems

- Workability
- Carbonation

Percieved problems

- Workability, good properties can be controlled by admixtures
- New admixtures now available
- Good cohesion, well suited for SCC

Limestone Calcined Clay Cement LC3

LOW CARBON LOW COST LOW CAPITAL

Self compacting concrete: cohesion

50% FA: 1.5% WRA

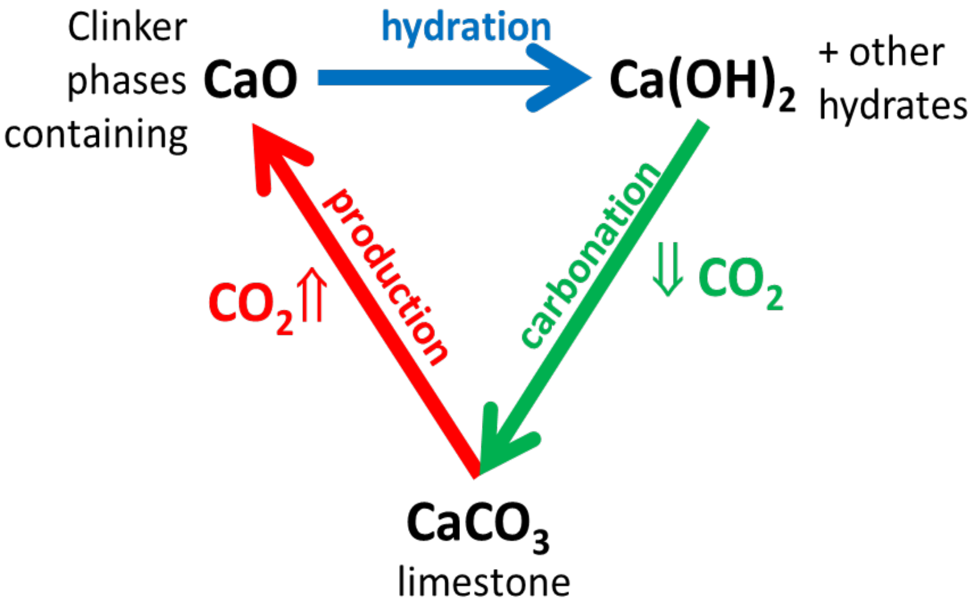
50% LC^2: 1.2% WRA

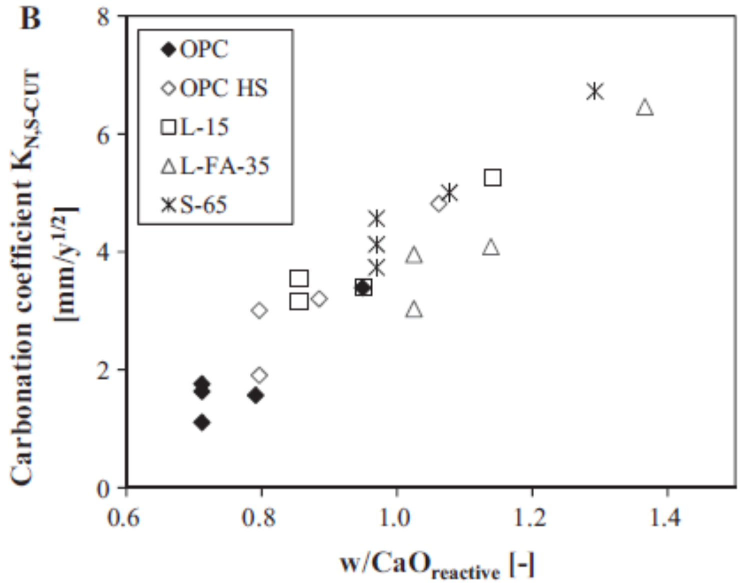
Harsh Vardhan et al. 2020

EPFL

Good quality low-tech concrete

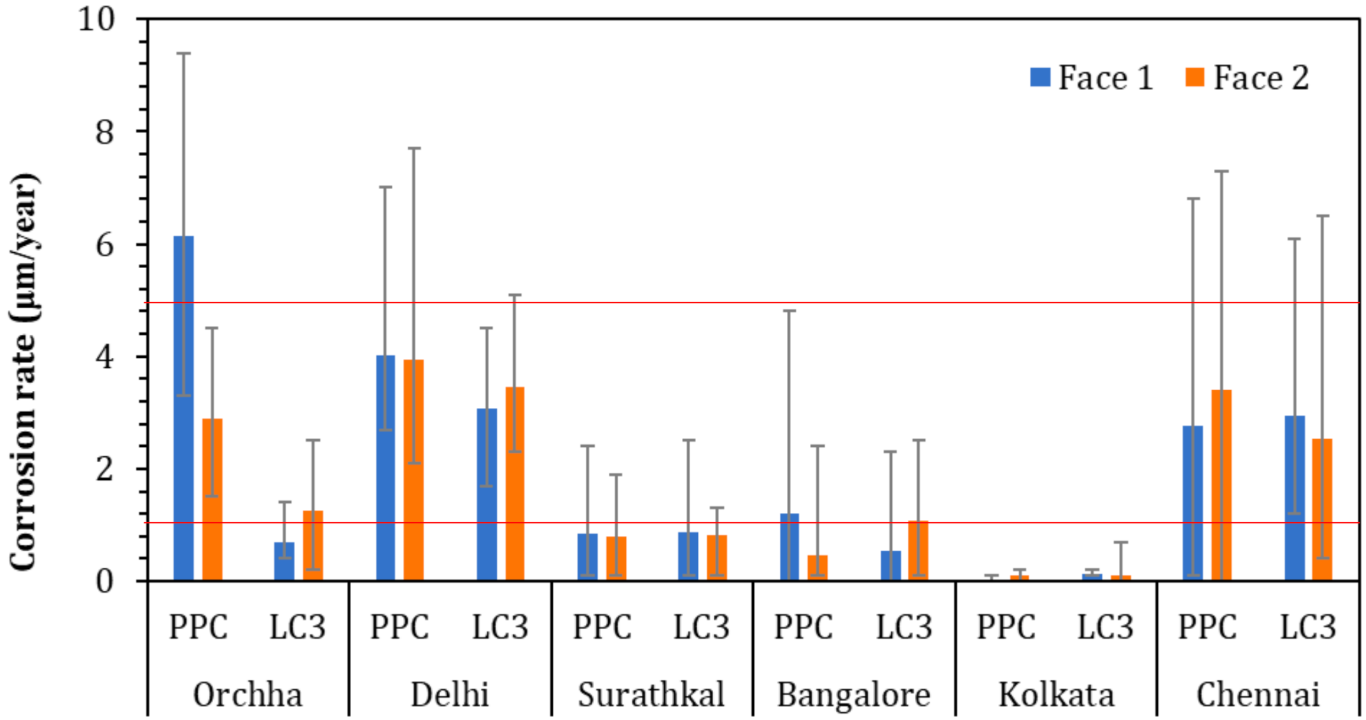
All blended cements will carbonate faster, but carbonation rates will still give service life > 50-100 years





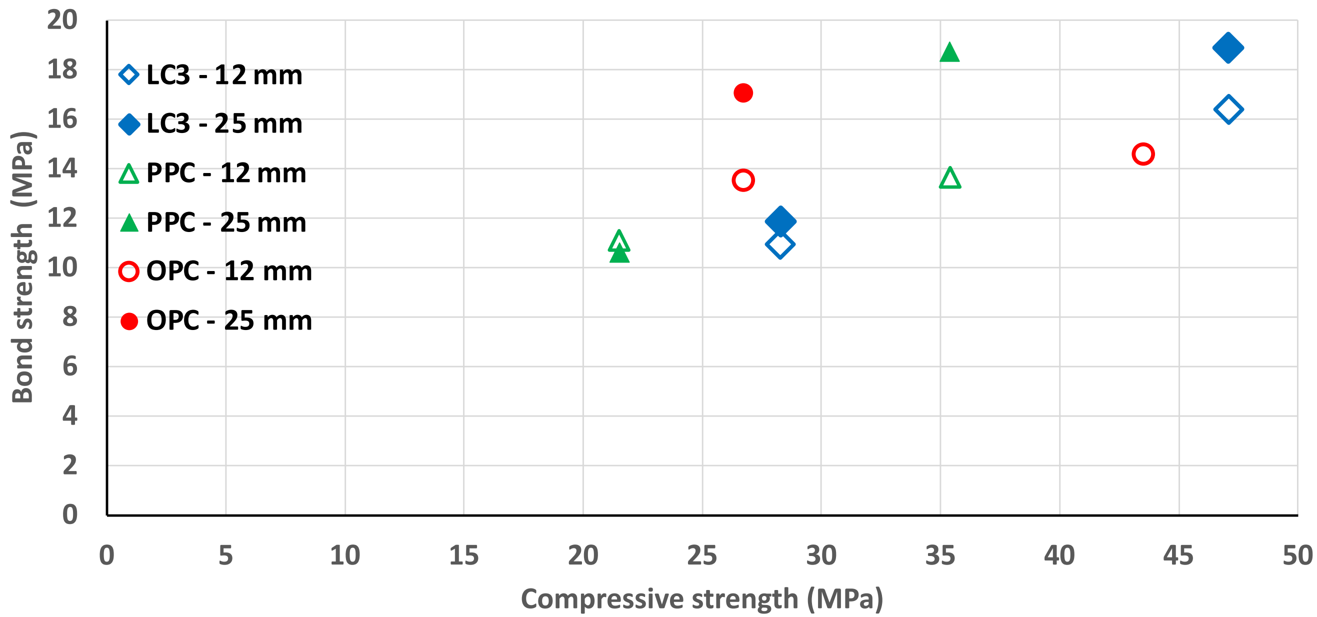
Field data on carbonation

Field data on carbonation

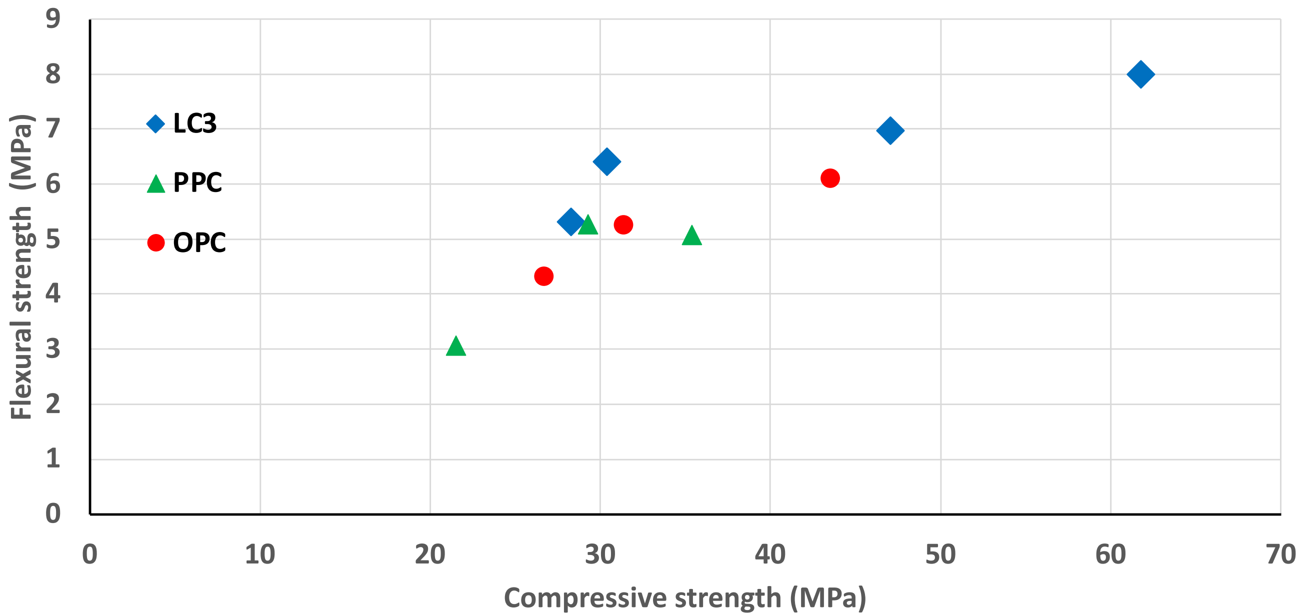


Impact Engineering properties

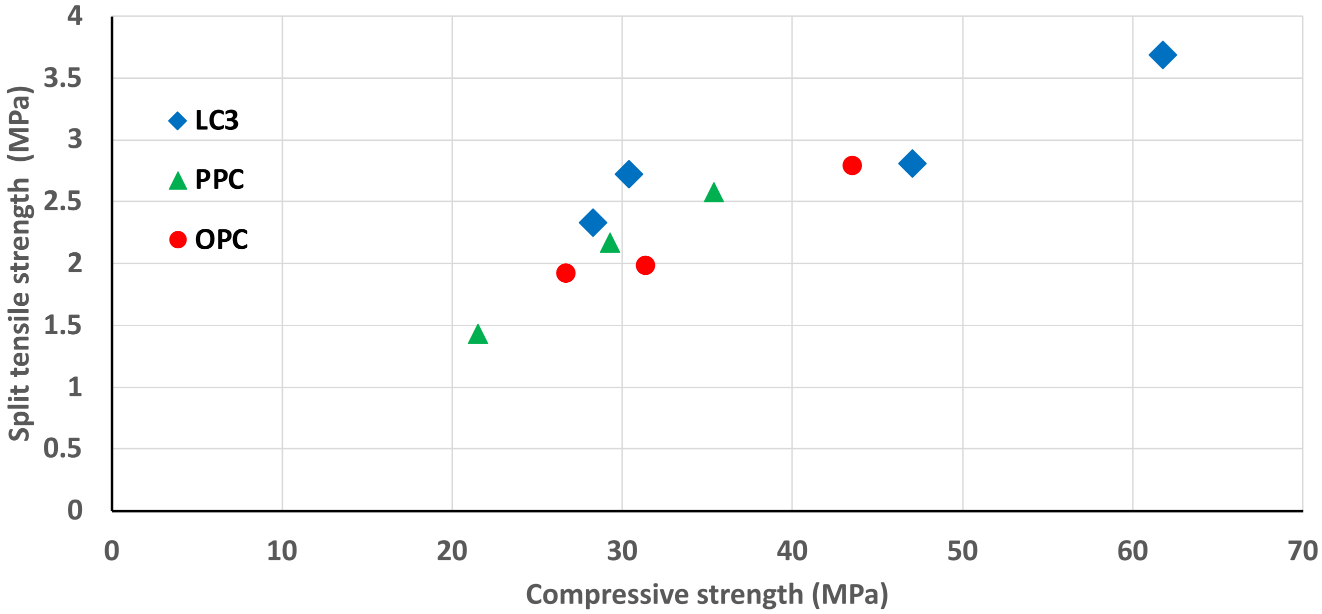
Bond with reinforcement



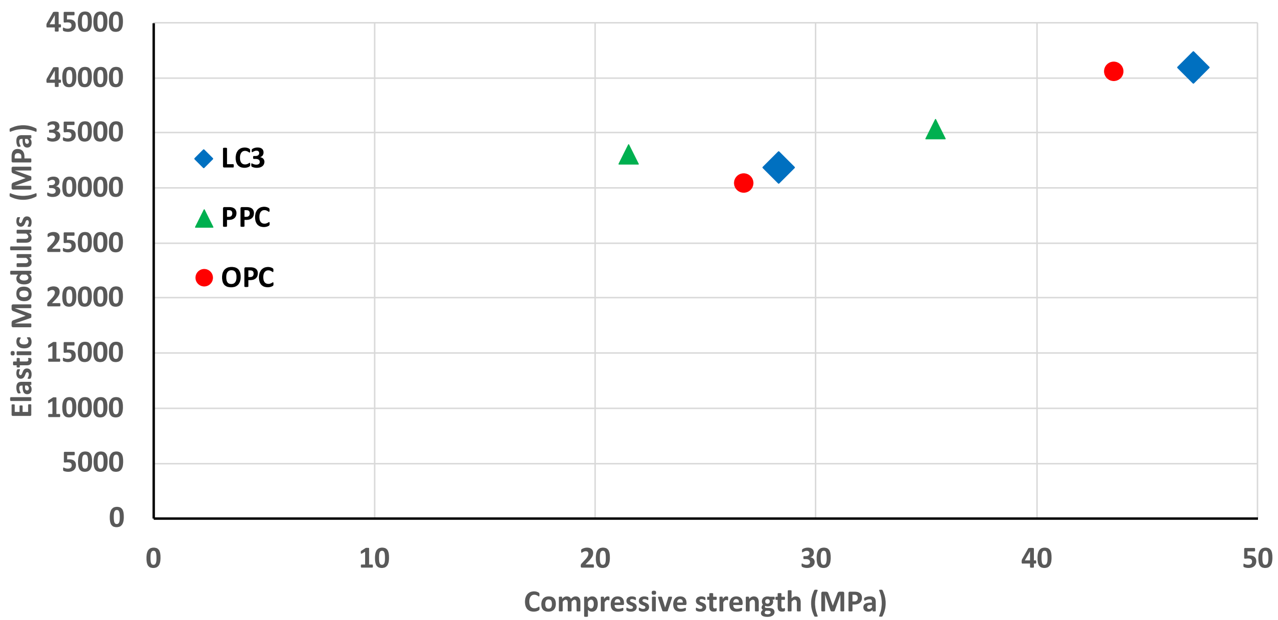
Flexural strength



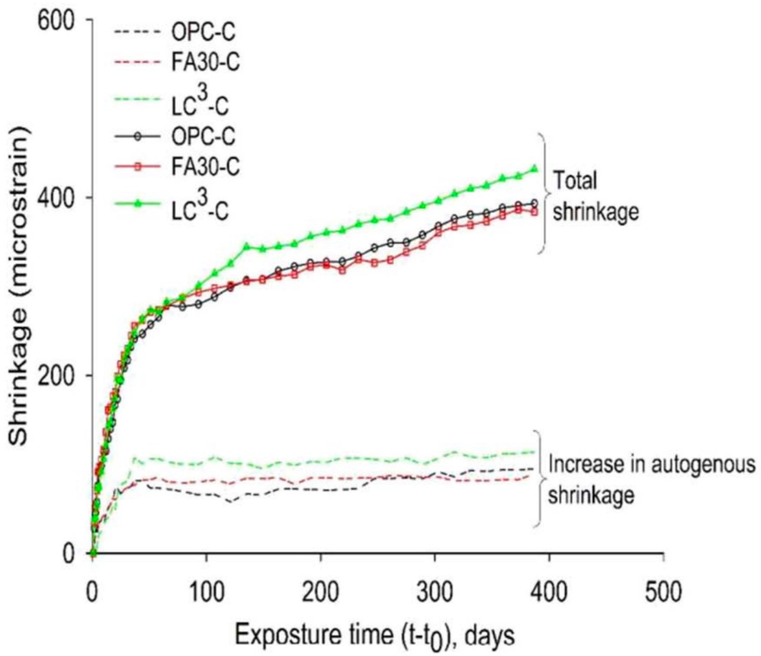
Split tensile strength

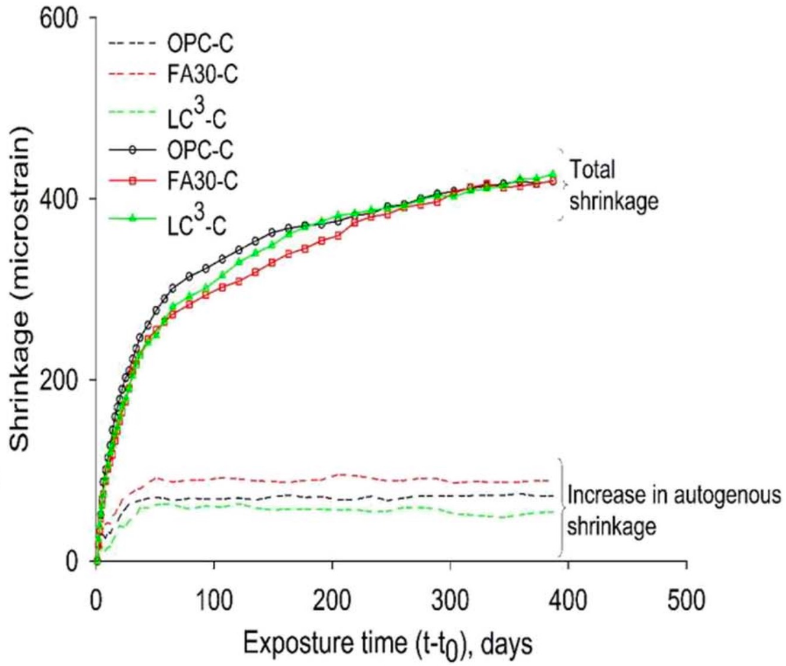


Elastic Modulus



Shrinkage of concrete



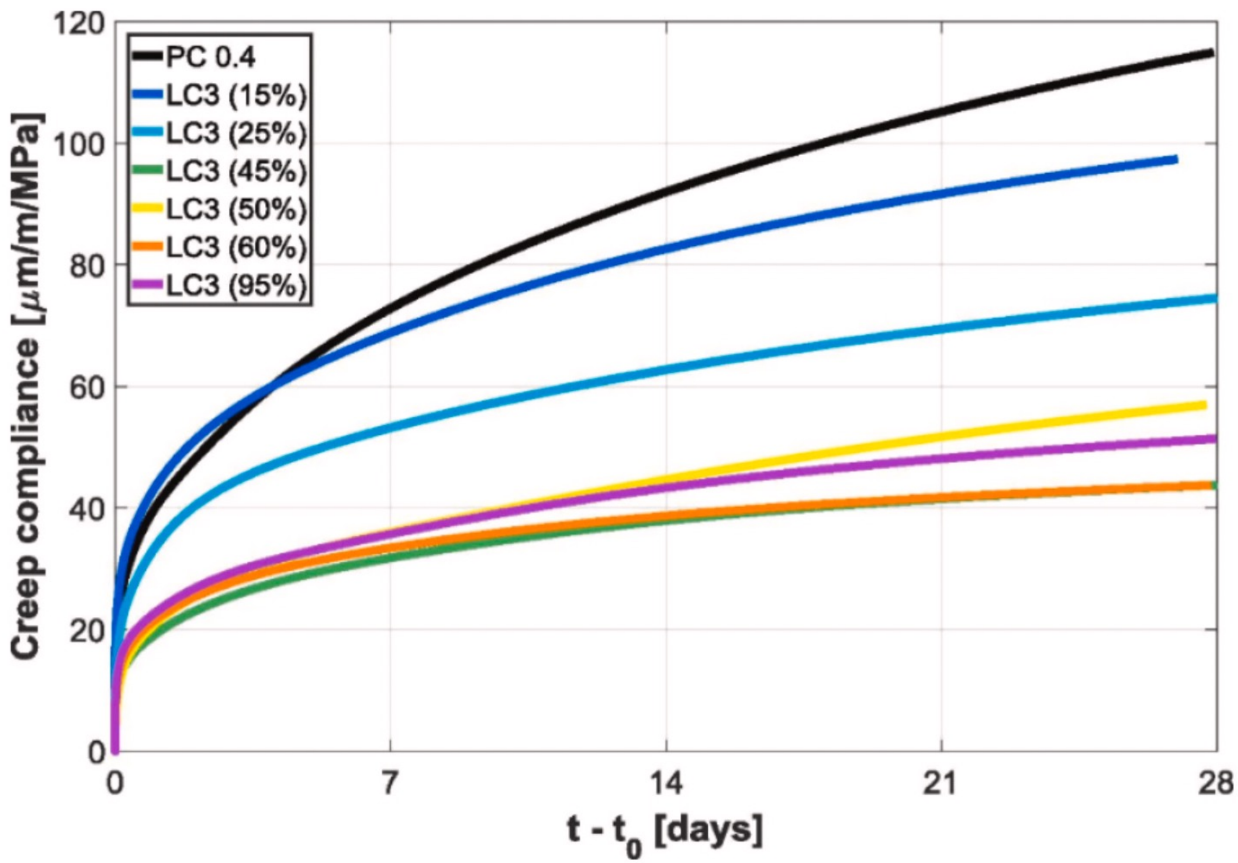


Dhandapani at al. 2018

Limestone Calcined Clay Cement LC3

LOW CARBON LOW COST LOW CAPITAL

Creep, significantly lower



Ston et al. 2019

EPFL

Limestone Calcined Clay Cement LC3

LOW CARBON LOW COST LOW CAPITAL

Comparison of LC3 concrete with concretes prescribed in Dubai

A report on the Dubai Building Code for sustainable concrete - 2021 edition

ÉCOLE POLYTECHNIQUE FÉDÉRALE DE LAUSANNE

Objectives

Dubai Building Code

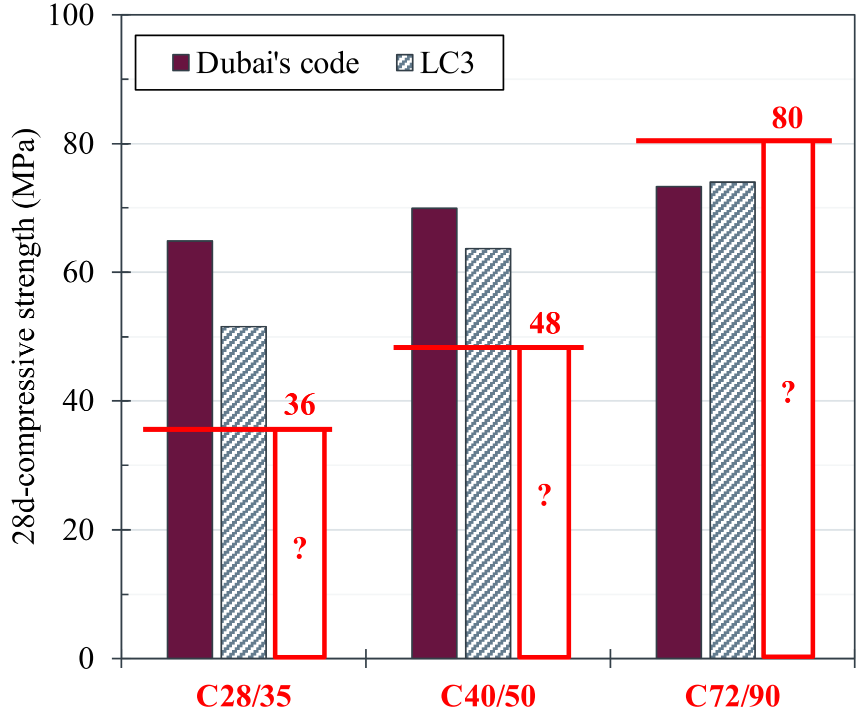
Validate the performance of the mixes prescribed

Dubai vs. LC³ 50

Compare the carbon footprint and strength by simple replacement

LC3-optimized
My contribution

- Reduce the binder content
- Match the code’s requirement
- Improve the consistency.



GWP database

Materials

OPC was Holcim Normo4

GGBS and SF from previous projects

Calcined Clay from Heidelberg

Limestone, gypsum and commercial pure calcium hydroxide

PCE based SP from Sika (ViscoCrete)

Performed tests

7 and 28d-compressive strength

28d-RCPT

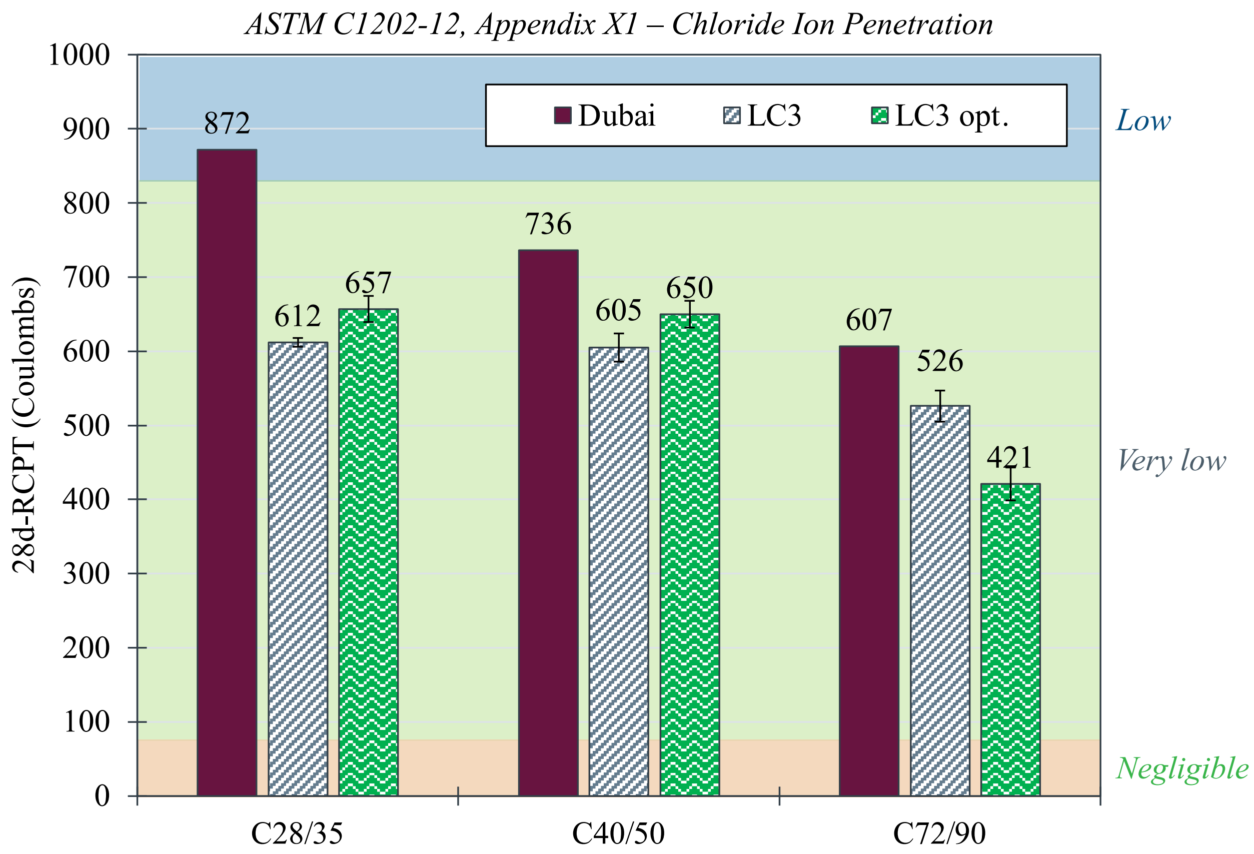
1y-natural carbonation

Our database

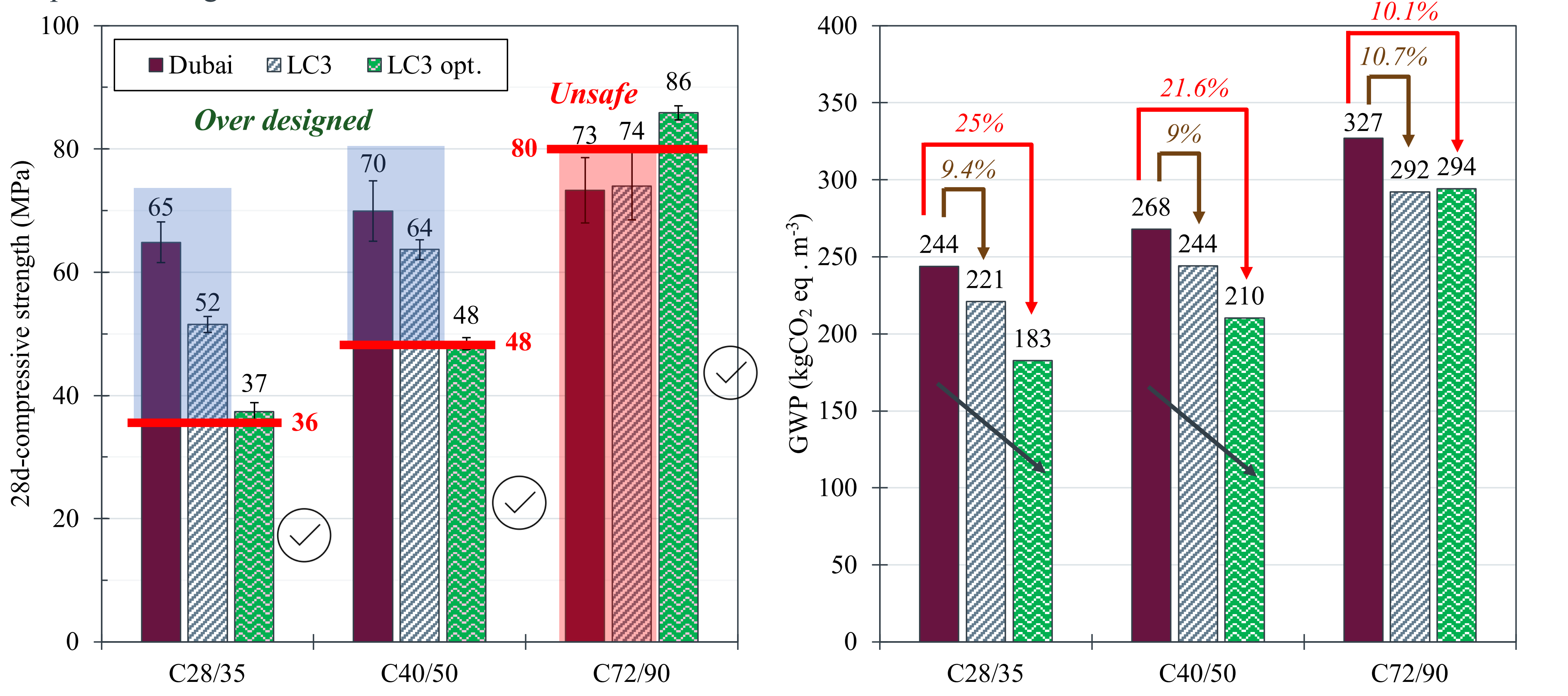
Materials	GWP (kgCO_2eq / tonne)
OPC	827
Slag	179
Silica Fume	134
Calcined clay	232
Limestone	48
Gypsum	54
Fine agg.	5
Coarse agg.	11
SP	1305

Mix designs

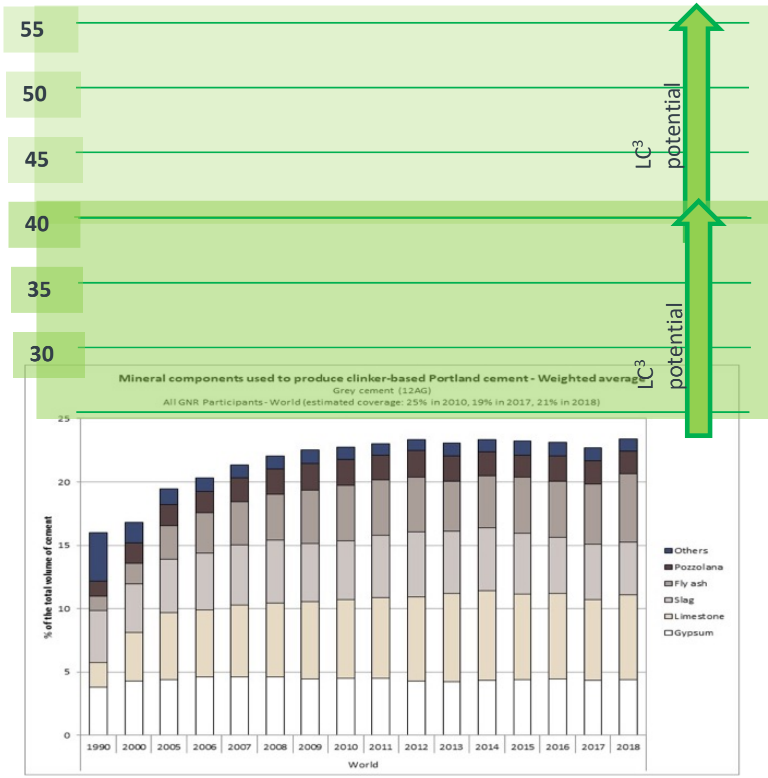
Strength class	C28/35			C40/50			C72/90		
Materials (kg/m³)	Dubai	LC³	LC³ opt.	Dubai	LC³	LC³ opt.	Dubai	LC³	LC³ opt.
Total binder	**380**	**380**	**325**	**420**	**420**	**375**	**510**	**510**	**510**
GGBS ratio	36%			36%			26%		
SF ratio		55kg (*15%*)			45kg (*11%*)		8%		
w/b ratio	**0.42**	**0.42**	**0.61**	**0.36**	**0.36**	**0.48**	**0.29**	**0.29**	**0.26**
SP (%)	0.50	1.56	0.20	0.50	1.97	0.50	0.75	1.97	2.50
Slump test (mm)	10	-	**100**	10	-	**75**	10	-	**10**


ASTM C1202-12, Appendix X1 – Chloride Ion Penetration
28d-RCPT (Coulombs)
1000
900
800
700
600
500
400
300
200
100
0
Dubai
LC3
LC3 opt.
872
612
657
736
605
650
607
526
421
C28/35
C40/50
C72/90
Low
Very low
Negligible

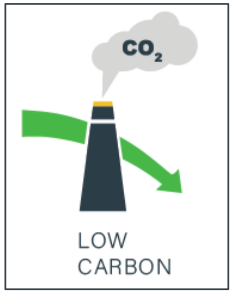
Compressive strength and GWP



Calcined Clay only SCM which can expand substitution

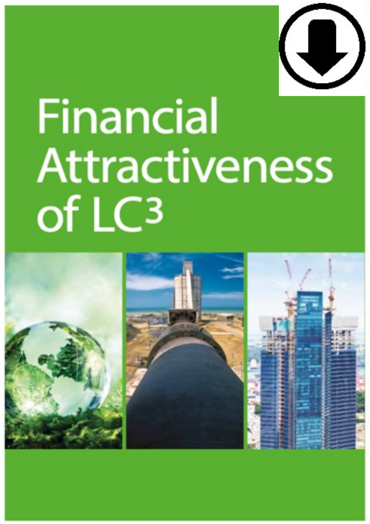


- ✓ 800 million tonnes CO_2/yr
- ✓ 400 million tonnes CO_2/yr



EPFL LMC

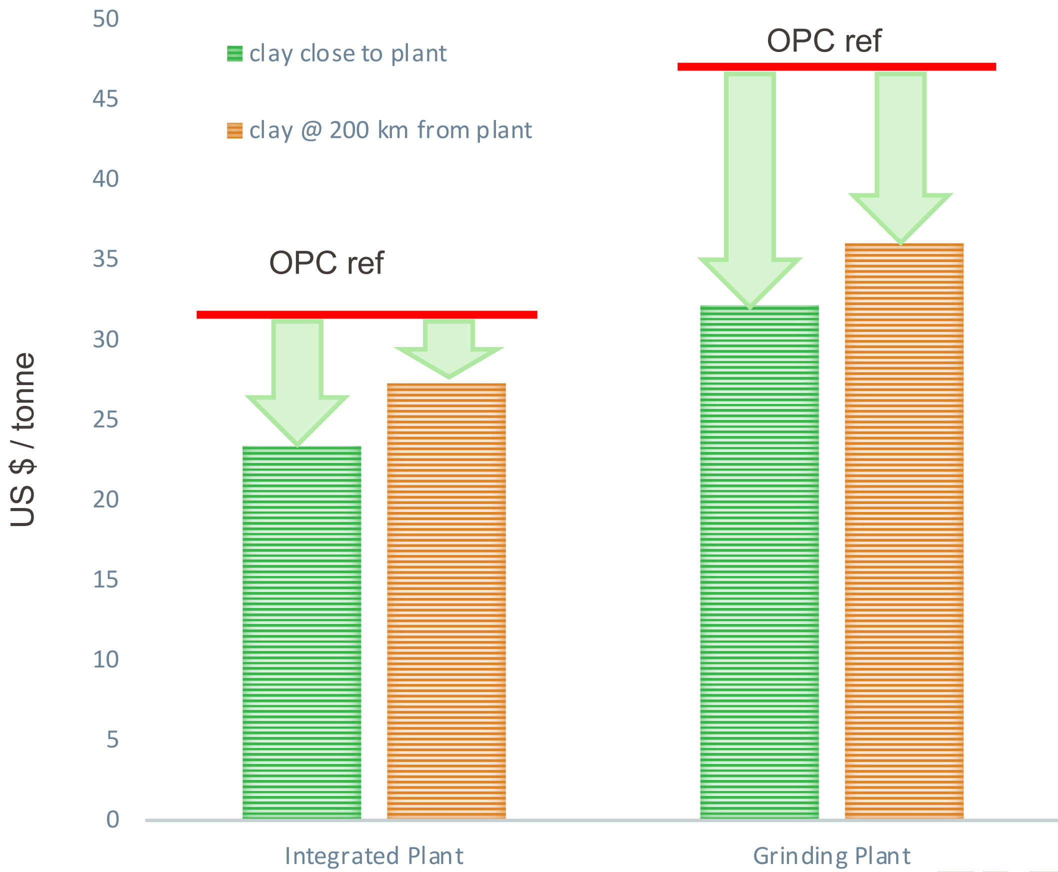
Financial Feasibility



Report available to download:
www.lc3.ch

Study by LC3 Project partner CEMENTIS

LOW
CAPITAL



LC2 treatment in standards: based on UK situation

Prof. F. Martirena

2023-08-05

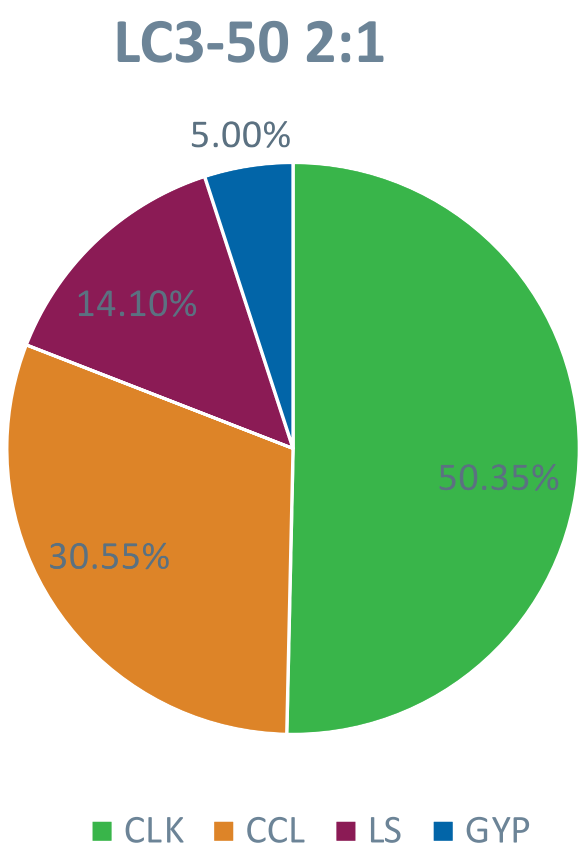
LC3 designation according to EN-197-1 (2020)

- Portland-composite cement EN 197-5 – CEM II/C-M (Q-LL)
  - Composition:
    - Clinker (K) 50%
    - Limestone (LL) between 15-20%
    - Calcined pozzolana (Q) between 30-35%
  - Denominations:
    - CEM II/C-M (Q-LL) 32,5 N (or R)
    - CEM II/C-M (Q-LL) 42,5 N (or R)
    - CEM II/C-M (Q-LL) 52,5 N (or R)
- Options for the production of this cement (on the cement factory)
  - Co-grinding all components together (K-Q-LL)
  - Separate grinding
    - CEM I 52,5 N (or lower strength)
    - LC2 (60% Q-35%LL-5% gypsum)
    - CEM I and LC2 will be blended and homogenized at plant
    - C2 (95%Q+5% gypsum)
    - CEM II
  - Both cements must comply with composition of EN 197-5 – CEM II/C-M (Q-LL)

Options for using calcined clays-limestone as cement extender

» LC2: Blending 50% of pure Portland cement (CEM I) with 50% of a blend of calcined clay (60%), limestone (35%) and gypsum (5%), that is called “LC2

» C2: Blending 70% of Portland Limestone cement (PLC) with 30% of sulphated calcined clay (95% calcined clay and 5% gypsum)



Limestone Calcined Clay Cement LC3

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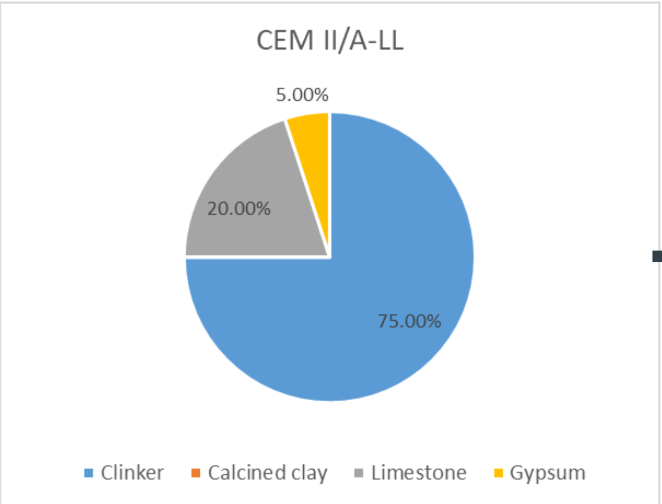
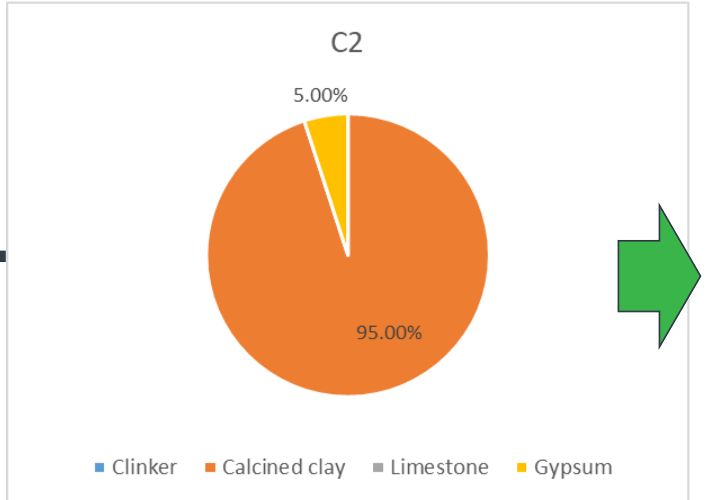
The combination of CEM II/A-LL and sulphated calcined clay (C2) for LC3-50

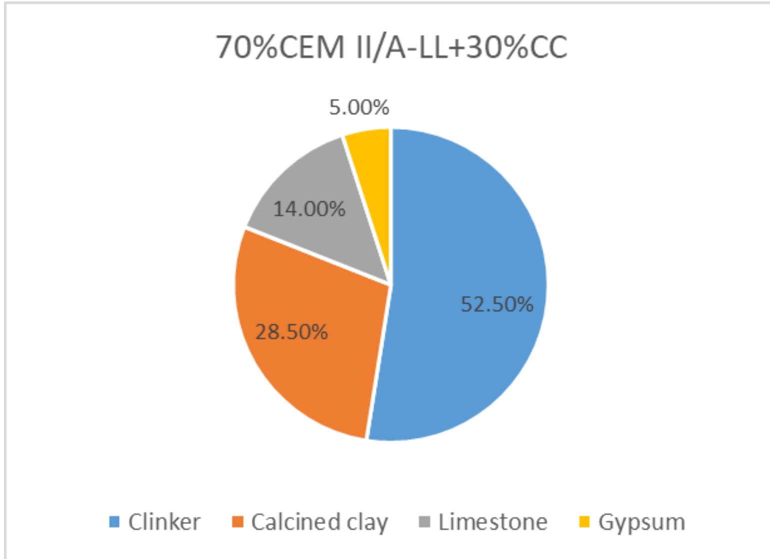


70%

+



30%



This option is already available and covered by BS standards

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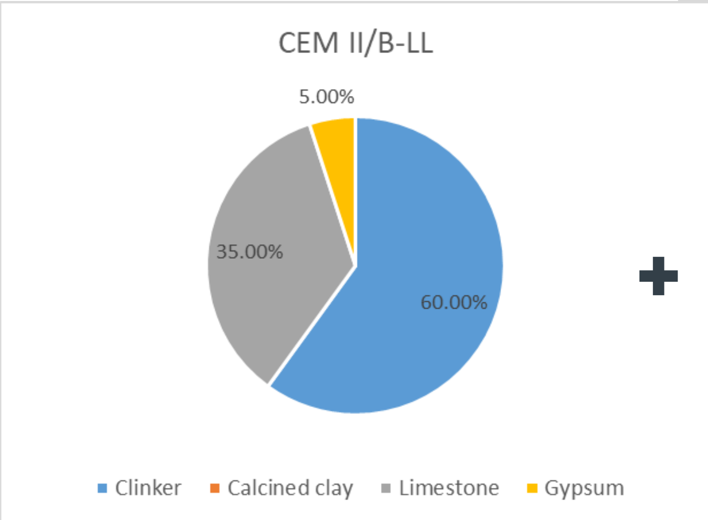
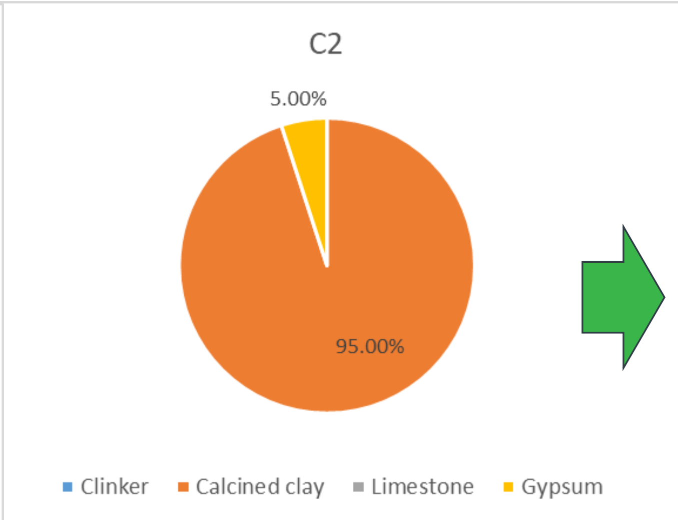
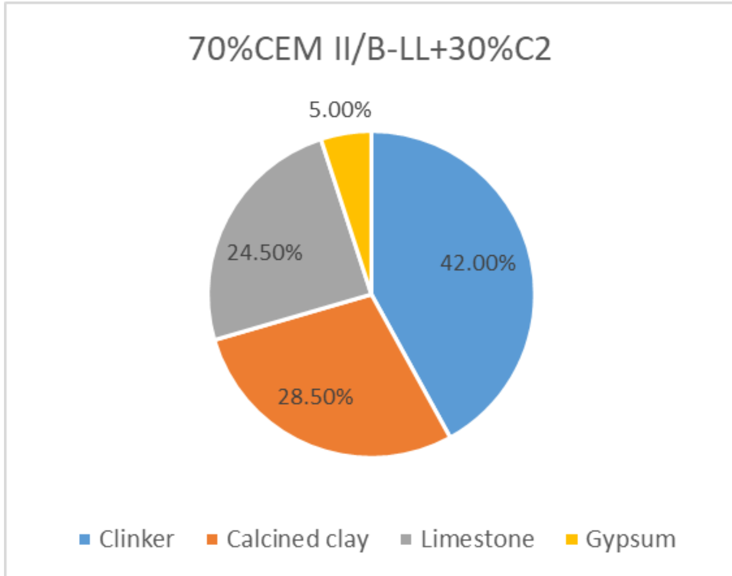
The combination of CEM II/B-LL) and sulphated calcined clay (C2) for LC3-40







70% **30%**

This option is already available and covered by BS standards

Limestone Calcined Clay Cement LC3

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Options for standards with C2

BSI committee B/517/4 has agreed, inter alia, to “correct” an earlier omission to allow Calcium Sulfate (gypsum) to be added to calcined clay in BS 8615.

Correction 1 to BS 8615-1

4.3.7

Replace ‘BS EN 196-2’ with ‘BS EN 196-2 as modified by 4.3.1’.

4.3.8

Replace ‘BS EN 196-2’ with ‘BS EN 196-2 as modified by 4.3.1’.

4.3.10

Replace ‘BS EN 196-2’ with ‘BS EN 196-2 as modified by 4.3.1’.

Correction 1 to BS 8615-2

4.3.8

Replace ‘BS EN 196-2’ with ‘BS EN 196-2 as modified by 4.3.1’.

4.3.9

Replace ‘BS EN 196-2’ with ‘BS EN 196-2 as modified by 4.3.1’.

4.3.11

Replace ‘BS EN 196-2’ with ‘BS EN 196-2 as modified by 4.3.1’.

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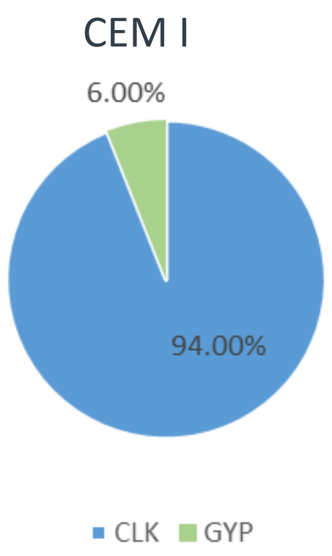
Options for standards with C2

BSI committee B/517/4 has agreed, inter alia, to "correct" an earlier omission to allow Calcium Sulfate (gypsum) to be added to calcined clay in BS 8615.

Amendment 1 to BS 8615-2
Add a new 4.3.14
4.3.14 Calcium sulfate
Calcium sulfate may be added to the other constituents of calcined pozzolana during its manufacture to control setting when the addition is mixed with cement and water.
Calcium sulfate can be gypsum (calcium sulfate dihydrate, $CaSO_4 \cdot 2H_2O$), hemihydrate ($Ca_SO_4 \cdot$ ½H2O), or anhydrite (anhydrous calcium sulfate, $CaSO_4$) or any mixture of them. Gypsum and anhydrite are found naturally. Calcium sulfate is also available as a by-product of certain industrial processes.
4.3.5 Add a new second paragraph
Where the natural calcined pozzolana comprises calcined clay, it is permitted to add sulfate so that **the sulfate content shall not be greater than a mass fraction of 3.0% of the calcined clay**.

Options for standards with LC2



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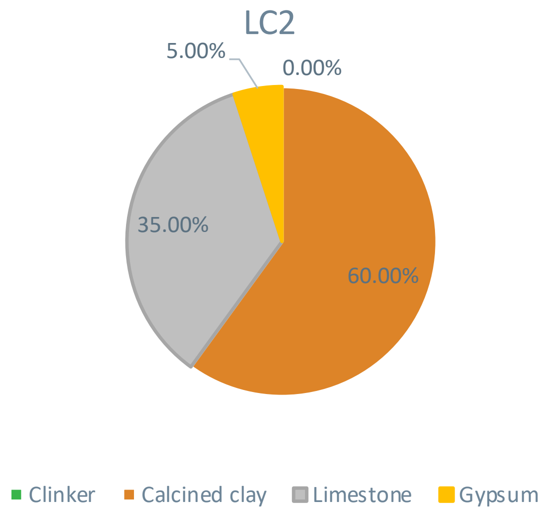

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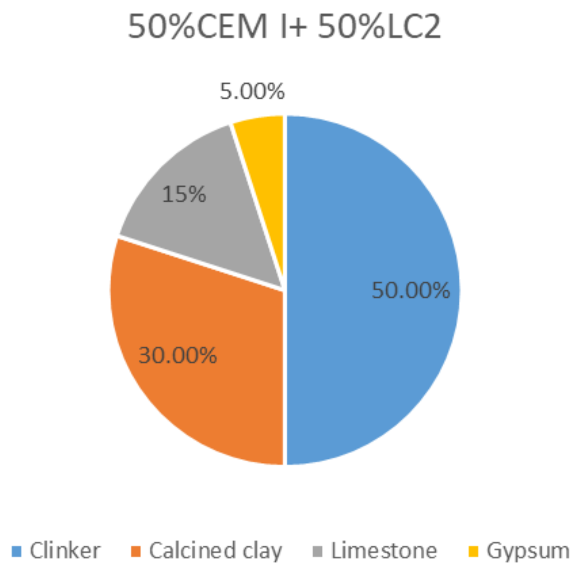


50%CEM I + 50%LC2

CLK: Clinker
GYP: gypsum
CCL: calcined clay
LS: limestone

The GREAT advantage of LC2 is that it can be added flexibly based on final use; it also has a long shelf life

Options for standards with LC2

- LC2 would be marketed as 60% CC + 40% Limestone + added sulfate (gypsum) and be blended at the readymix plant 50/50 with CEMI (42.5R or 52.5N or R). The resultant blend would have an optimal sulfate content.
- The cement industry has successfully resisted (through BS EN 15167-1 and BS 8615-2:2019) the addition of sulfate to ground slag and calcined clay.
- Currently a CEMI 42.5R must have a maximum SO_3 content of 4.5%. This means that a combined 50/50 BS 8615-2 LC2 + CEMI would have a relatively low SO_3 content, impacting concrete performance.
- Market access could be achieved by applying for and obtaining a Publicly Available Specification (PAS) see - https://www.bsigroup.com/en-GB/our-services/developing-new-standards/Develop-a-PAS/PAS-consultancy-enquiry-form/brochure/.
- PASs can be drafted/sponsored by the producer and submitted to the relevant steering committee set up by BSI and representing all interests. (Interestingly, one geopolymer producer has adopted this route with PAS 8820.2016)

Publicly Available Specification (PAS)

- » It could comprise (1) a product comprising calcined clay (conforming to BS 8615) plus sulfate (gypsum) and (2) an LC2 comprising calcined clay and limestone (conforming to BS 7979) with the permission to add sulfate. In both cases, the resultant sulfate content of the blend expressed as SO_3 by mass of addition would be required not to exceed the limit for the combination strength class, but the resultant blend would be able to benefit from optimal sulfate contents.
- » A PAS for LC2 does not completely address the liability issue for specifiers and project owners, but can provide a kitemark for the product and arguably opens up the market for less challenging concrete applications (see LC3-50 narrative above).
- » It would typically take 12 to 15 months from the start of a project to a published PAS. For a sponsored PAS the costs would be about £120k. Adding sulfate to an addition is likely to be controversial but is believed to be winnable.
- BS 8500 uses the abbreviation II/C-M to permit both a CEMII/C-M cement or a mixer combination of cement plus addition to be supplied and treated as being the same.
- » The interim result would allow both a calcined clay + gypsum and a calcined clay + limestone + gypsum product to be sold directly to readymixers and concrete product manufacturers, to create either a CII/B-M or CII/C-M.

Interim options for the use of LC2 before PAS is approved

- Ready mix plants often have a silo for very fine limestone to be used as mineral addition. LS should comply with BS 7979
- They would have to build up a second silo for C2
- Each SCM (C2 and LS) could be added to the mixer together with cement (CEM I or CEM II/A-LL or CEM II/B-LL and finally have an LC3 cement on the mix with varying clinker content.
- The restrictions for the use of the blend would be:
  - Minimum cement content according to BS EN206 and BS8500
  - Some exposure classes that according to BS EN 206 and BS 8500 limit the amount of mineral additions in concrete (or need to be further proved). Tests include resistance to carbonation, chlorides, freeze-thaw, and sulfate environments, as well as susceptibility to alkali silica reaction and use in concretes containing steel and other reinforcement.

Concluding remarks

- ✓ Substantial reductions in CO_2 are possible
  - ✓ At cement level by increasing SCM substitution
  - ✓ At concrete level by minimising cement content
  - ✓ At structure level
- ✓ All of the above will also lower cost
- ✓ Remainder CO_2 can only be dealt with by carbon capture and storage at a high cost, infrastructure not in place.
- ✓ Calcined clays are the only realistic option for extending the use SCMs
- ✓ Can be done FAST and at SCALE

Limestone Calcined Clay Cement LC3

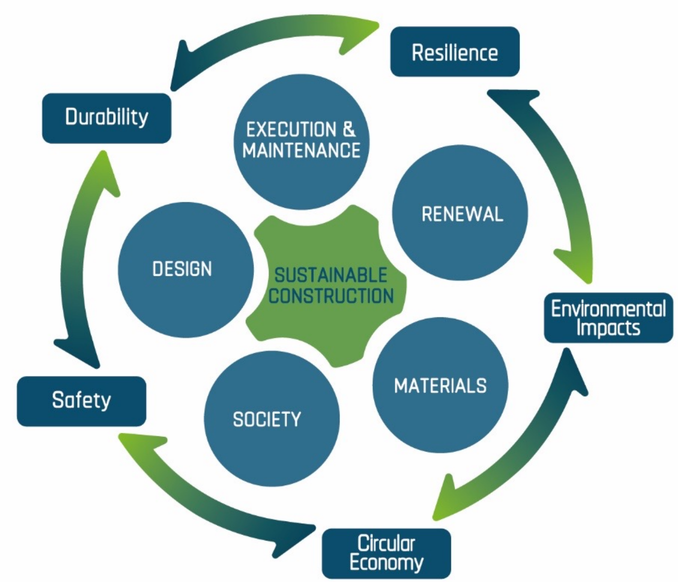
LOW CARBON LOW COST LOW CAPITAL

Recommending a new paradigm for the built environment

GLOBE

Global consensus on sustainability in the built environment

- High level policy advice
- More than 150 nations
- 5000+ experts
- 50+ years of expert networks
- Standards and guidelines
- Research and education
- Innovation



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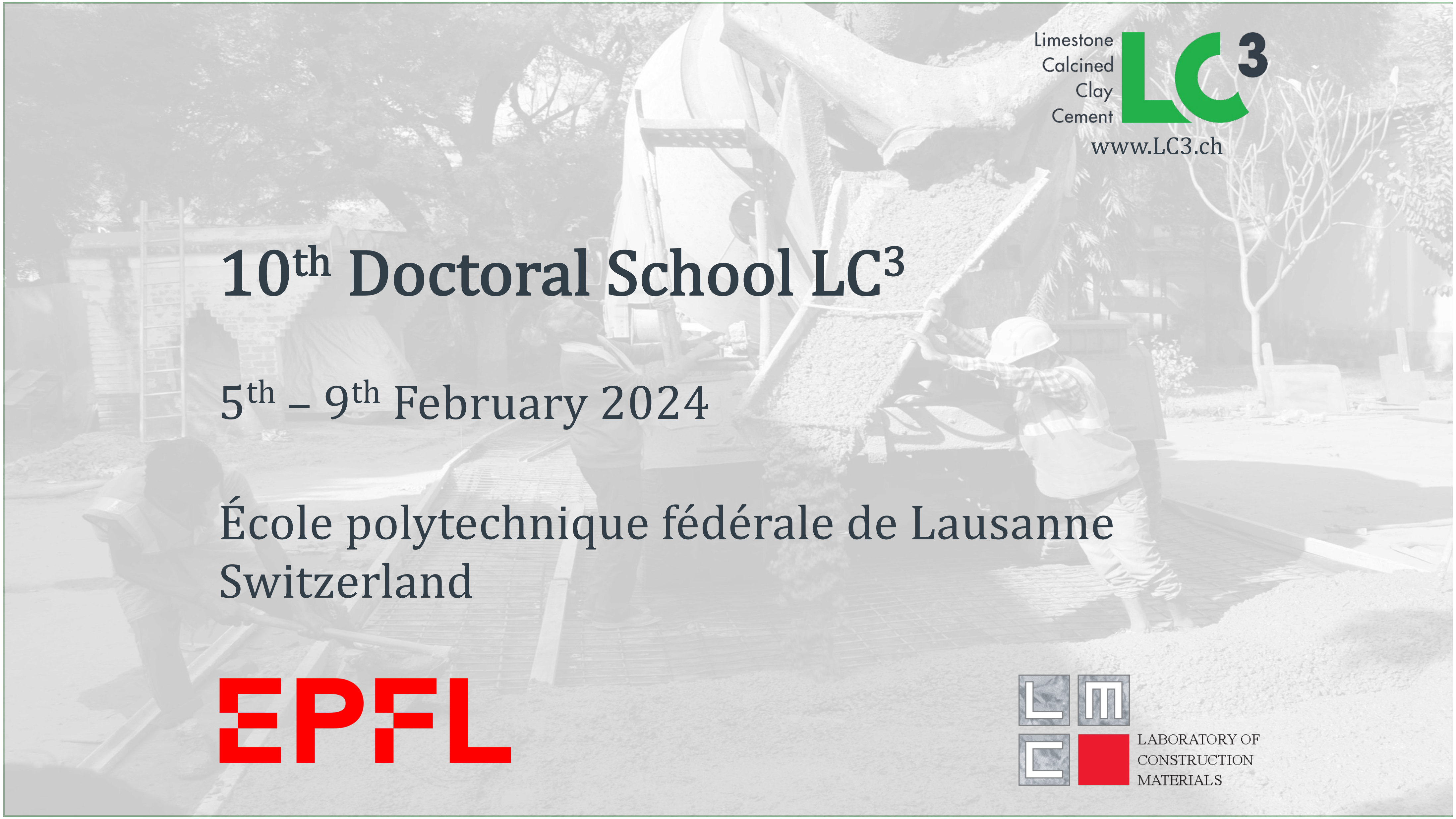

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15-18 May 2024

4th International Conference on Calcined Clays for Sustainable Concrete

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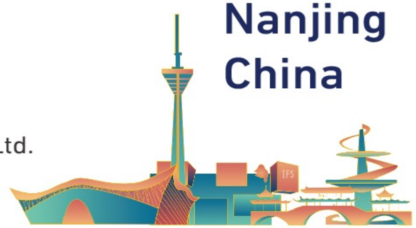

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Thank you

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World limestone deposits