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Eco-efficient cements: No magic bullet needed

Against the increasingly urgent need to reduce industrial CO_2 emissions, there has been significant research into alternative cement chemistries that claim drastic reductions. However, the geological reality of the elements available from the earth mean there will be no magic solutions, although there are still lots of opportunities for improvement...



Above: Karen Scrivener is the head of the Laboratory of Construction Materials at Ecole Polytechnique Fédérale de Lausanne (EPFL), Switzerland.

Right - Figure 1: Relative CO₂ and embodied energy intensity of building materials, relative to virgin aluminium (the highest).

ement is critical to the built environment and will continue to be a major factor in construction over the coming decades. This is because, despite some recent bad press, concrete, the 'real' final product of the cement sector, has no comparable substitute at present. Indeed, concrete has very favourable embodied CO2 compared to other materials (See Figure 1). Not only does it come out well compared to steel, aluminium and glass, it even scores well against wood, which is riding something of a wave at the moment on the back of its apparent 'green' credentials. Even as it currently stands though, at just 15% of the size of the cement sector, wood is an unsustainable proposition. Many more forests are being cut down than planted. It cannot, in any meaningful manner, dent demand for cement and concrete.

That said, the cement and concrete sectors cannot be complacent. The world is set to overshoot the CO₂ emissions that will cause more than 1.5° C of warming compared to pre-industrial levels by 2028. Clearly we are going to overshoot and, when that happens, we will then have to remove CO₂ from the atmosphere, in addition to rolling-out largescale industrial carbon capture and storage (CCS), reforesting large areas of the earth's surface and a host of other remedial efforts. Therefore, anything we can do now to mitigate the rise in temperature is extemely valuable, not just in terms of the environment but also in terms of what we'll have to spend on fixing problems later.

The importance of concrete will not change in the future, if anything it will become more critical. As global populations grow, particularly in Africa and the Far East, urbanisation will continue to accelerate, driving further demand for cement and concrete to provide a decent standard of living. If we restrict concrete production to decrease CO_2 emissions, the world will face increased pressure from mass migration.

Ways to reduce cement CO₂ emissions

The cement sector clearly has an important role to play in CO_2 mitigation efforts and major steps have



already been made. Alternative fuels, particularly biomass, are one approach, as are supplementary cementitious materials (SCMs).

Looking ahead, CCS, which might be a cost that could be tolerated in the west, is currently economically prohibitive in developing markets. Also, some say that 50% of the emissions reduction in the cement sector could be achieved by so-called 'innovative solutions.' Unfortunately, the geological reality of what's actually available within the earth makes many of these propositions unsuitable for large-scale use. Figure 2 shows the options: Just eight elements: oxygen, silicon, aluminium, iron, calcium, sodium, potassium and magnesium account for 98% of the earth's crust.

Clearly for a material to be used in the same quantities as cement and concrete, it needs to be based entirely on these eight elements, simply because there are insufficient quantities of everything else. This is a constraint but it also means that we don't have to exhaustively study all possible options. We have to study the options that are available.

Straight off the bat, we can discount cement systems based on sodium and potassium. Their oxides are far too soluble to ever deposit hydrates in a hypothetical cement system. On the other end of the

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scale, magnesium and iron are not soluble enough. They can sit in concrete structures for decades and still fail to contribute to strength development.

So, from eight elements, we're down to just three abundant oxides, those of calcium, aluminium and silicon. Within these possible systems, there are then only two main compositions that can be used for hydraulic cements: Calcium silicates (Portland cement) and calcium aluminates. The latter are of interest from a CO_2 mitigation standpoint, since they contain far less CaO. This means less limestone and less CO_2 than with Portland cements.

Unfortunately things are not as simple as we might like, as to make calcium aluminate cements, we need materials that have high proportions of aluminium and relatively low levels of silicon. These are not widely distributed on earth. 90% of bauxite, for example, is concentrated in just 10 countries. Baux-



ite is also expensive, because it is used primarily for the production of aluminium. Even if we took all the bauxite and dedicated it exclusively to calcium aluminate cement, we'd only be able to produce 10-15% of the current global cement demand. Calcium aluminated cements thus remain interesting for certain applications, but they won't ever replace Portland cement in a meaningful way.

From the above process of elimination, it is clear that continued production of large quantities of Portland cement production is inevitable. The sector has incredible economies of scale and the materials needed are available everywhere. The reaction kinetics are ideal for construction. We don't have any material that can do the same job.

Increased use of blended cements

In the absence of suitable alternative materials and with CCS nascent at best, the most practical answer to lowering CO_2 emissions from cement and concrete prodution is by extending the use of blended cements. This is by far the most effective lever to reduce cement-related CO_2 emissions and enhance sustainability. However, of late SCM use has reached something of a plateau. Two of the three main SCMs, slag and fly ash, are limited in supply and are becoming increasingly scarce. This is estimated to limit the potential reduction in global clinker factor to around 70% if only these materials are considered.

Let's look at slag first. It is a fantastic SCM and you can easily make blends using as little as 30% clinker. Globally though, slag production is only 8% of what would be needed keep up with that level. Of that amount, 95% is already used in cement or concrete, so there really is no further potential for slag to dramatically reduce CO_2 emissions from cement and concrete.

Slightly more available at present, but not in a CO_2 -constrained future, is fly ash. If we're really serious about tackling CO_2 emissions, then fly ash will be the first SCM to disappear. We have to stop burning coal because it represents 60% of the world's CO_2 emissions.

Of course we have very large quantities of limestone but after 15% addition of limestone we're basically just diluting the clinker. There is some potential for increased proportions of limestone but it's hard to see this as a major solution in itself. Other SCMs are out there: Vegetable ashes are available in small quantities, for example, and natural pozzolans may be the answer for some users in some locations. However, the volumes are not forthcoming.

Calcined clays provide opportunities

Without new SCMs it won't be possible to push the global clinker factor much below 70%. Thankfully, calcined clays provide an answer. They are widely available and could enable reduction to an average clinker factor of 50% of even as low as 40% for some

Left - Figure 2: The relative abundance of elements in the earth's crust, by number of atoms.

Left: New approaches like CCS are unlikely to come to bear in developing markets for some considerable time. A better approach is to provide practical solutions that make use of existing technologies and skills.



Right: Discarded clays could provide a valuable new source of SCMs, as shown at this Indian quarry.

formulations. What is particuarly interesting is the use of calcined clays with clinker and limestone. This approach is being investigated by the LC3 project, an EPFL-led project supported by the Swiss Agency for Development and Cooperation that began in 2013.

It has long been known that calcined clays can be pozzolanic. When used alone, the maximum substitution level is around 30%. This gives a moderate saving in CO_2 emissions. However, if we substitute a further 15% of the clinker with limestone, we get a significant reduction in CO_2 emissions, with a product that has almost identical properties to the blend containing just the calcined clay.

Strength results for Portland cement and an LC3 blend with only 50% clinker are comparable, even showing higher strength for the LC3 blend after seven days. So, the blend uses 50% less clinker, produces 30-40% less CO₂ and offers similar strength

to Portland Cement. If scaled up worldwide, this would lead to reductions of several hundred of million of tonnes of CO_2 per year, equivalent to those of a country the size of France. Some properties, notably resistance to chloride, are also significantly improved.

Why are calcined clays so reactive?

Clays contain Kaolin, which is formed of alternating layers of silica and alumina. When you heat it up to 750-800°C the silica and alumina layers are disrupted and can then both react. When you add limestone too, the alumina contained within the meta-kaolin reacts with the limestone to give space-filling hydrates, particularly mono and hemicarboaluminate. These form to a limited extent in limestone cements but the reaction can go much further in the LC3 blends.



Left: Demonstration house in India made using LC3 cement.

Below: Demonstration house in Santa Clara, Cuba made using LC3 cement.



Right - Figure 3: Comparison of mortars made with plain OPC or binary blends with 30% replacement of calcined clay, slag or fly ash and ternary blends each with an additional 15% limestone.





The LC3 project has compared the strength development for various binary and ternary systems (Figure 3). Already at four days the calcined clay systems are well ahead of slag and fly ash. The existing limits on blended cements are related to early strength, so this is a great improvement over the binary mixes. It can be used like Portland or blended cement, like-for-like. No supplementary equipment or training is required.





Where are the clays?

Kaolinitic clays are available in a large number of countries, particularly in Africa, Asia and Latin America, where demand for cement is likely to increase most strongly. Even outside of these, there are quality clays to use. The LC3 project has been working in Rajasthan in India, not renowned for its clays, but there is still abundant material. Indeed, many of the possible feedstocks have already been discarded from other production processes. Chinese LC3 collaborators have identified 3Bnt of 'waste' material perfectly suitable for making LC3 cement containing 50% kaolin at a single site.

The state of play

Full scale production trials of LC3 cement have already taken place around the world, particularly in Cuba and India. Processes have been developed and example structures have been built. The first commerical production of LC3 cement is due to begin in Latin America in the first half of 2020.

Concluding remarks

LC3 cements are only one part of the solution to the problem of cement and concrete CO_2 emissions. Not only do we have to look at the clinker factor in cement, but also the 'cement factor' in concrete and the concrete intensity of the built environment. If each of those can be minimised, we can take great strides towards much less CO_2 intensive construction processes.

By looking at metrics like CO_2 intensity as a function of compressive strength, we can find better ways to build using concrete. It makes sense to move away from site mixing to centralised mixing, as this provides greater control over the amount of cement used. Indeed, a shift towards higher use of pre-cast components should be encouraged for the same reasons. The amount of cement wasted also needs to be looked at, particularly arising from bagged cement. China, for example, has now banned the use of bagged cement in certain major municipalities. This is estimated to offer emissions reduction of 5% in those municipalities, just by reducing of the amount of material lost.

Finally to come full circle, as the title states, there are no 'magic bullets' for reducing cement-related CO_2 emissions. However, by working throughout the value chain CO_2 emissions could be reduced by 80% compared to 1990, without huge extra costs, all while using existing knowledge and codes.

Right: Production of LC3 blocks by KJS Concrete in Dadri, India.

Right: Ripped bags containing discarded and unusable cement abound on the world's construction sites. This waste comes with a CO₂ cost and efforts should be made to reduce such instances.