BEDP ENVIRONMENT DESIGN GUIDE

LOW CO₂ CONCRETE: ARE WE MAKING ANY PROGRESS?

Peter Duxson and John Provis

Summary of

ACTIONS TOWARDS SUSTAINABLE OUTCOMES

Environmental Issues/Principal Impacts

- Construction materials contribute 5-8 per cent of all global anthropogenic CO₂ emissions, and this figure will increase in the
 coming decades as the developing world continues to use more and more concrete.
- The predominant source of CO₂ in the construction industry is the calcination of limestone and silica to produce Portland cement. There are limited possibilities for environmental advances in this process due to the fundamental chemistry of the raw materials used.
- Alternatives to Portland cement exist, and include alkali-activated binders, which utilise industrial wastes such as blast furnace slag and coal fly ash to produce a cement-free concrete.
- · Prescriptive concrete standards are constraining innovation, though performance-based standards are being developed.

Basic Strategies

In many design situations, boundaries and constraints limit the application of cutting EDGe actions. In these circumstances, designers should at least consider the following:

- Be aware of the high CO₂ emissions required in the production of concrete.
- Consider alternative binders for concrete where appropriate.
- Consider maximising re-use of wastes, such as aggregates, where appropriate, and where likely to produce net positive
 environmental gains. Note that reuse of some materials may require significant energy to process.

Cutting EDGe Strategies

- Scientifically selected mixtures incorporating waste materials for instance alkali activated binders may be preferred to ad
 hoc blending of wastes with cement.
- As standards for their use are yet to be developed, be aware use of such materials in non-structural applications until such time as a standards regime is in place.
- Participation in standards development should become a priority for all stakeholders including end-users, producers and
 other interested parties to ensure that the documents produced are of maximum value.

Synergies and References

- RILEM Technical Committee on Alkali Activated Materials, http://www.rilem.net/tcDetails.php?tc=AAM
- BEDP Environment Design Guide: PRO 7: The Environmental Impact of Building Materials
- BEDP Environment Design Guide: PRO 31: Concrete and Sustainability Supporting Environmentally Responsible Decision Making

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While concrete is the most widely used building material in the world with many architectural and engineering benefits, it is associated with a high environmental cost that somewhat offsets this benefit. Previous EDG papers have investigated measures that can be taken to improve the environmental performance of concrete, with a brief introduction to emerging products that can actually replace traditional Ordinary Portland Cement in concrete.

This paper provides an explanation of the existing market forces, and an update on the emergence of alternative low carbon dioxide (CO_2) cements and the role they can play in the future of concrete construction

Kevwords

alkali activation, alternative cements, carbon dioxide (CO2), concrete, standards, sustainable development

1.0 INTRODUCTION

Modern cement and concrete technology has provided the backbone of much of the world's construction since its development in Europe in the 1800s. Concrete is produced by blending a 'binder' (usually cement and water) with aggregates (usually sand and gravel), to produce a thick slurry which is then cast or poured into the shape of its final form and allowed to cure. During curing it hardens, gains strength and gives off heat, and reaches its design strength in a period of a few weeks.

Conventional concrete made from Ordinary Portland Cement (OPC) is second only to water as the commodity most used by mankind today. Global OPC production in 2008 is estimated to be 2.6 billion tons per annum in a thorough market analysis (Freedonia Group, 2008), while other sources (Cement Sustainability Initiative, 2007) mention 1.5 billion tons per annum, which is a huge variation. This \$150 billion industry (Freedonia Group, 2008) contributes conservatively 5-8 per cent of global (CO₂) emissions (Scrivener and Kirkpatrick, 2008), mainly as a result of the decomposition of limestone during the energy intensive cement making process. With rapid development in infrastructure in China, India, the Middle East and elsewhere in Asia, the cement and concrete industries are expected to expand significantly. Cement usage in Australia is relatively stable at approximately 13-15 megatonnes, with organic growth only. During the recent 'bull' (or rising) market and commodities boom, shortages of cement supply became apparent in Australia and in many locations around the world. While this has been impacted on slightly as a result of the 2008 global economic slowdown, demand from the developing world in particular is expected to remain strong. As a result of established infrastructure, market dominance and no significant driver to use alternative binders, OPC is currently accepted technically and commercially as the only viable binder to make concrete, even if Supplementary Cementitious Materials (SCM; materials other than cement, which will be discussed in detail in this article) comprise between 10-60 per cent of the binder.

Alternative binder systems, such as alkali activated cements (which will be the primary focus of the

discussion here), supersulfated cements, or phosphatebased, and magnesium-based cements, despite presenting a significant reduction in CO2 emissions, have not been taken up on a commercial scale as CO2 has not historically been a significant driver for new technologies. Usually the driver for competition has been either cost reduction, in which case new materials starting from a low volume basis can never compete against large scale OPC production, or significantly superior performance. In terms of concrete, even products utilising OPC with significant performance benefits (i.e. high strength/durability concrete) have found it very difficult to penetrate the market due to increased cost, reinforcing the fact that the short-term cost implications of concrete are clearly the dominant factor.

It has been five years since the EDG explored the topic of alternative binders for concrete in the paper $PRO\ 31$: Concrete and sustainability – Supporting environmentally responsible decision making. (Hes and Bates, 2003), at which time alternative cements were not yet commercially available in Australia. This paper explores the existing use of OPC and SCM in concrete and their implications in terms of environmental footprint, and provides a market update and progress report on the emergence of alternative low CO_2 cements over the past five years, the barriers to their market uptake, and the role they can play in the future of construction.

2.0 EMBODIED ENERGY IN CONCRETE

2.1 Ordinary Portland Cement

Ordinary Portland Cement (OPC) is the grey powder that is mixed with water, rock and sand to create concrete. Cement is the largest commodity product on the planet next to water. The term 'cement' identifies 100 per cent OPC, although recently the cement industry has been using the term to describe a blend of OPC, fly ash (a waste product of coal combustion) and slag (a waste product of iron and steelmaking) which are the SCM referred to above. OPC is made of a complex mixture of calcium silicates, aluminates, aluminoferrites

and sulfates, which is most easily described in terms of its oxide composition. Refer to box below.

The source of calcium is limestone, which is mainly calcium carbonate (CaCO₃), and it is obtained through mining. The other elements are obtained from clays, shales and other sources. To make cement, these minerals are heated at approximately 1400°C, during which time CO_2 is released from the calcium carbonate (CaCO₃) and fusion of the materials occurs to produce 5-20mm balls of hard grey material called 'clinker'. This process releases around 0.50-0.55 tonnes CO_2 per tonne of clinker generated, via the chemical reaction calcium carbonate (CaCO₃) \rightarrow calcium oxide + CO_2 ("calcination"), and approximately 0.40 tonnes CO_2 is produced from the fossil fuels used to generate the energy used to heat the materials to 1400°C.

This results in a material with approximately 1 tonne CO₂ emissions for every 1 tonne of cement.

2.2 Influence of Sand/ Aggregate, Recycled Aggregate and Concrete

Despite aggregate and sand accounting for approximately 30 per cent of all emissions during the production of concrete, recycling concrete or aggregate creates few opportunities to reduce carbon emissions (Cement Sustainability Initiative, 2008). Green House Gas (GHG) emission reductions can be obtained when a high carbon footprint material or process is substituted for a lower one. Recycling concrete into aggregate, or different aggregate sources into aggregate for concrete, tends not to produce any such savings compared to using natural aggregate except insofar as transportation requirements can be reduced.

Cement manufacture is the target area for carbon emission reductions as it is the stage of production where most GHG impact occurs, and some tentative but slow and incremental steps have been made by the industry as a whole in recent years (World Business Council for Sustainable Development, 2008). Despite this, recycled aggregate has been a focus of the concrete industry, as there is no solution to the fundamental CO_2 emissions related to OPC production and in this way the industry can 'be seen to be doing something'. Unfortunately, these efforts appear to be actually achieving little in real terms on the most important measure – CO_2 emissions.

Most recycled concrete is used as aggregate, although the strength of the original concrete needs to at least match

that required of the new concrete. Well cleaned recycled coarse aggregate can be comparable in quality to virgin aggregate, although this is generally not considered not a commercial or practicable practice. Due to contamination and costs of processing, little recycled material is generally used in concrete for anything other than footpaths, kerbs and gutters, and this does not result in a large sustainable reduction in virgin material usage or CO₂ emissions.

Europe is probably the most advanced market for recycled aggregate and yet it accounts for only approximately 6-8 per cent of the use of these materials. There is very limited capacity for recycled aggregate to play any significant role in reducing the environmental impact of concrete, since so little product is recycled compared to production. For instance, all the available recycled aggregate in Australia does not provide enough material to fill Australia's basic needs for road base.

A study by the American National Ready Mix Concrete Association (NRMCA) has concluded that up to 10 per cent recycled concrete aggregate is suitable as a substitute for virgin aggregate for most concrete applications, including structural concrete (Obla et al. 2007). UK research recommends that up to 20 per cent of recycled concrete aggregate can be used for most applications (including structural) (Dhir & Paine, 2007). Australian guidelines state that up to 30 per cent recycled aggregate can be used for structural concrete without any noticeable difference in workability and strength compared with natural aggregate (Clarke et al. 2008). Nonetheless, the difficulties experienced during the construction of Council House 2 for Melbourne City Council (Melbourne City Council 2006) suggest that this may be overstating the case somewhat.

There remains significant potential for an increase in the use of coarse recycled aggregate in concrete, though one should ask whether this will reduce emissions or the use of non-virgin materials over the entire economy? It may be that a lower class but widely distributed use such as road base could have more environmental benefitial impact than if recycled aggregate is only used in selected prestigious building projects such as Council House 2.

3.0 ALTERNATIVES TO OPC

If the use of recycled aggregates is neither possible on any substantial scale nor capable of reducing CO₂ emissions of concrete – are there any alternatives to OPC? Detailed surveys of the different technologies available for GHG emission reduction in construction materials have been provided recently by Gartner (2004) and by Phair (2006); a brief summary of some of the relevant technologies and issues is presented here.

Oxide composition of Ordinary Portland Cement

50-60 % calcium oxide (CaO), commonly known as burnt lime, lime or quicklime

30-40 % silicon dioxide (SiO₂), commonly known as silica

remainder aluminium oxide (alumina, Al₂O₃)

iron oxide (Fe₂O₃) sulfur trioxide (SO₃)

UEPG 2006 statistics published 2008 have a figure of 6 per cent. QPA (October 2007) has higher figures and gives 2006 statistics as 8 per cent European average and 26 per cent in Great Britain.



Figure 1. Concrete with geopolymer binder being used in footpath construction in Hume City Council, Victoria

(Source: Courtesy of Zeobond)

3.1 OPC Blends using Supplementary Cementitious Materials

Probably the most straightforward and technologically accessible way to reduce the GHG emissions associated with cement production is simply to use less cement in concrete. This is most commonly achieved by blending the cement with one or more pozzolanic materials, defined as materials which can react with the calcium hydroxide (Ca(OH)2) formed during cement hydration to produce cementitious binding phases. These are also referred to as Supplementary Cementitious Materials, or SCMs. Commonly used SCMs include blast furnace slag, coal fly ash, silica fume (all produced as industrial wastes), calcined clays, and volcanic ash. Each of these materials has a much lower embodied CO2 content than Portland cement, and with arguably zero CO₂ emissions attributable to them, as these materials would otherwise have been discarded to landfill (including most fly ashes and slags). In addition to enhancing the sustainability of blended cements, appropriately selected SCMs can also bring performance advantages including enhanced resistance to acid attack, decreased permeability and modified workability (Bouzoubaâ et al. 1999; Bilodeau and Malhotra, 2000; Shi and Qian 2000; Sabir et al. 2001).

As an additional advantage, blended cements generally possess similar rheology and chemistry to standard Portland cements, meaning that the skills base and standards regime required for their use are already largely in place. However, the lower reactivity of many but not all SCMs, when compared with cement, can cause difficulties in early strength development, which limits the maximum achievable substitution in many applications. This also places a cap on the GHG savings which may be achieved, given that a certain percentage

of the binder must still be comprised of Portland cement – often around 70-80 per cent, although blends with as little as 30 per cent cement have been used in some applications depending on performance requirements.

3.2 Magnesia-based Cements

One of the first developed alternatives to calcium silicate-based cements is magnesia cement, which in its most fundamental incarnation is based on the formation of magnesium oxychloride binding phases and is named Sorel cement after its inventor (Sorel 1867). Unfortunately, Sorel cement suffers from the chemical instability of several of the key binding phases when exposed to water or acid (Maravelaki-Kalaitzaki and Moraitou 1999), although various researchers are working to remedy this by the use of chemical additives or by the use of sulfates as well as chlorides in the synthesis process.

Calcination of magnesium carbonate (MgCO₃) to form magnesium oxide, or magnesia (MgO), takes place several hundred degrees below the temperature required for the decomposition of calcium carbonate (CaCO₃), giving some savings in fuel-derived CO₂ emissions (Phair 2006). However, due entirely to the very high percentage CO₂ by mass of magnesium carbonate (where CO₂ makes up more than 50 per cent of its total mass), the CO₂ emitted per tonne of magnesium oxide produced is markedly higher than is the case for calcium carbonate. Work is underway to develop magnesia cements whose chemistry is tailored to minimise this problem, by enhancing the CO₂ uptake of the hardened products. This means that much of the CO₂ emitted during production is then re-absorbed over the lifespan of the material. The Eco-Cements marketed by Australian company TecEco (TecEco, 2008) are based on this principle, as well as the use of

efficient furnace and grinding technology to minimise emissions during calcination.

Magnesium phosphate chemistry has also been utilised in forming cements (including 'Ceramicrete'), predominantly for niche applications such as nuclear waste treatment or oil well cements (Wagh, 2004), and display very high early strength. However, high raw materials and production costs mean that large-scale Portland cement replacement seems unlikely until the price of energy and emissions increases the price of OPC to approximately double current levels.

3.3 Sulfoaluminate Cements

Cements based on calcium sulfoaluminate chemistry have been utilised for a number of decades in Europe and China on a scale of more than a million tonnes per year for low heat and high durability. Sulfoaluminate cements are starting to see wider application worldwide as their advantages in various areas are observed (Glasser and Zhang, 2001). The binding phase in these materials is generally based on ettringite and related compounds, and clinker formation takes place at significantly lower temperatures and with markedly less process-related CO₂ emission than is attributed to Portland cement formation. This is due in large part to the lower calcium oxide content of sulfoaluminate cements compared to Portland cement, including the use of belite (ß-dicalcium silicate) rather than alite (tricalcium silicate) as a primary calcium silicate source.

The expansiveness of some of the phases in this system can sometimes be problematic, and carbonation may also be an issue. However, given that the production of 'Klein's compound' ($4\text{CaO} \cdot 3\text{Al}_2\text{O}_3 \cdot \text{SO}_3$), the predominant phase in a sulfoaluminate clinker produces less than half the CO_2 of alite (Gartner, 2004), there does seem to be significant potential for Portland cement replacement in applications where expansion can be controlled (or is desirable). In reality, though, sulfoaluminate cements are made from blast furnace slag, making their long-term scalability to replace OPC on a meaningful level limited due to availability issues and the fact that slag is generally controlled by cement companies. This may explain the investment of OPC manufacturing companies in this technology.

3.4 Alkali-Activated Cements

Alkaline activation of blast furnace slag, other metallurgical slags and coal fly ash for use in construction materials has been the subject of investigation since the 1940s (Purdon, 1940), but commercialisation efforts to date have generally been sporadic. Alkaline activation refers to the process of blending the solid precursors with an alkaline solution (instead of the water used to hydrate Portland cement), which accelerates their reactions and enables a solid concrete to form within an acceptable timeframe. In the mid-1950s, Prof. Victor Glukhovsky of Kiev, Ukraine, investigated the binders used in ancient Roman and Egyptian structures (Glukhovsky, 1994). Based on these observations he developed binders called 'soil-cement', or 'geopolymers' combining aluminosilicate wastes such as slag with alkaline industrial waste

solutions. In the 1960s, when there was a shortage of Portland cement in the former Soviet Union, the Kiev team was involved in the construction of apartment buildings, railway sleepers, road sections, pipes, drainage and irrigation channels, flooring for dairy farms, pre-cast slabs and blocks, using alkali activated blast furnace slag (Shi et al. 2006). Subsequent studies have shown that these structures have high durability (Xu et al. 2008). It is relevant to note that the earlier work by the Kiev team was all based on slag and that their work on fly ash is more recent. A vast number of patents and standards were produced on the earlier slag mixes, but this documentation has been largely inaccessible to the West. The GHG savings achievable through the use of alkali-activated concrete have been estimated at around 80 per cent compared to Portland cement concretes (Duxson et al. 2007). Alkali-activated concrete technology has only recently (since 2006) been commercialised on a small but significant scale in Australia and internationally by Zeobond, who operate a small plant in Melbourne and also license their technology to domestic and international partners. This material, based on alkaline activation of blended fly ash and slag, has been proven through lifecycle analysis and laboratory testing to give environmental and performance advantages over Portland cement concrete. Most importantly, fly ash and slag are currently utilised in significant volumes by the cement and concrete industry, meaning that the necessary bulk solids handling facilities are already in place to make the product available competitively. The Centre for Sustainable Resource Processing (including partners Blue Circle Southern Cement, Rocla, and Golden Bay Cement) have also recently started down the path of alkali-activated materials. Though their efforts are currently at a demonstration scale at the moment, there is hope that commercial supply by these parties could

4.0 REMAINING BARRIERS TO THE UPTAKE OF ALTERNATIVE CEMENTS

Prescriptive Standards

commence in several years.

The existing standards for cement and concrete have been developed and refined effectively over the past century. In fact, the world's first quality control system was created by Joseph Bazelgette during the construction of the sewer system for London in 1859-1865, which was the first major project to use OPC. In solving problems of variability Bazelgette developed a system to control the water-to-cement ratio - a method which is still ingrained in the heart of both the EU and US standards - and one which does not necessarily have any meaning for alkali-activated or other alternative cements, because they do not always simply involve adding water to a solid precursor. Over the years these unofficial standards have become official, and now represent a system of market supply regulation rather than driving safety and maintaining technical performance.

The regulatory environment for cement and concrete has been fundamentally shaped by the bodies involved in cement and concrete manufacture, whereas the bodies that purchase the product and take the risk by insuring the product have traditionally been left out of this process despite the fact they are the only stakeholders who actually stand to lose. For example, many of the criteria written into cement standards exist for the purpose of controlling material flows, particularly restricting the ability to reduce cement content of concrete, or what is also known as clinker levels in cement. These standards are known as 'prescriptive' and are made regardless of technical performance. In continuously shifting the focus of standards towards performance and away from prescription, the door has opened to utilise binders that do not rely on OPC.

In Australia there are two types of cement: GP (General Purpose) which consists of at least 95 per cent OPC, and GB (General Blended) which contains more than 5 per cent fly ash and slag. The standards still prescribe 'some' OPC in both types, but clearly there is only a small step to be taken from GB towards a zero OPC cement. Nonetheless, anyone would be able to make an alkaliactivated or other alternative concrete that is able to fit within the existing Australian cement standards simply by adding a teaspoon of cement to each load of concrete.

Performance Standards

In fact, this shift towards truly performance-based standards was envisaged in the Appendix of Australian Standard AS3972 (Standards Australia, 2007): "For many years cement standards all over the world have been to a large degree prescriptive. Prescription-based specifications are convenient: the tests needed to police prescriptions are usually simple and quick to carry out. However this convenience is achieved at the expense of innovation and being able to easily incorporate new or advanced knowledge. With prescriptive specifications only a narrow range of solutions to any one problem is acceptable even though many other solutions may be available which would give equal or better performance." Further in the Appendix of AS3972, the basis for performance standards development is given: "The following three elements are essential for the development of a performance-based Standard:

- Performance parameter Usually the property or properties that best relate to the desired performance;
- Criteria Quality Level(s) of the required property that yield the desired performance;
- Test Method A clear, reliable, easy-to-use method of test which determines compliance with the criteria."

5.0 AUSTRALIA'S WORLD LEADING ROLE IN THE UPTAKE OF LOW CO₂ CONCRETE

Europe and China are the world leaders in sulfoaluminate cements, and the USA is the world leader in magnesium and phosphate cements. Australia is a world leader in alkali-activated materials, and customers are driving the uptake of innovative low-CO₂ solutions. (Eastern European nations while conducting a good deal of empirical mix-development work, has not focused on the progression of the scientific analysis of the technology.) Possibly due to greater environmental awareness, uptake in Australia has been particularly strong at the 'grass roots' in local councils, in large infrastructure projects and in private projects. While in the longer term as the product becomes more widely adopted, these drivers will reduce as all products improve environmentally, grass-roots support is a significant driver for uptake in the coming years while low-CO₂ concretes provide a massive reduction in emissions compared to OPC technology.

If one is not able to get engineering consulting firms to accept a new product, then it is immaterial whether or not the customer or architect specifies an alternative concrete product. Therefore, there is a leading role for engineers to play in driving the uptake of such a new material. Fortunately both consulting engineers and architects are more recently looking for environmental innovation.

6.0 CONCLUSIONS

Traditional cement, or OPC, is one of the most polluting products on the planet in terms of CO_2 emissions. While cement and concrete companies have implemented a number of mechanisms to reduce the CO_2 emissions of concrete, they have had only a very minimal effect. Use of SCMs and recycled aggregates does not avoid the inherent high emissions of OPC. Australia is already playing a leading role in developing alkali activation technology, one of the potential alternatives to Portland cement and which has the potential to reduce the CO_2 footprint of concrete by about 60 per cent.

There are other alternatives available in addition to alkali activation, including sulfoaluminate and magnesia-based cements, and different regions are leading the development of different types of technology. The main impediment facing the uptake of new construction materials is the existing standards regime, where prescriptive standards specify particular mix designs for concrete rather than allowing any material which meets given performance standards to be utilised. The international standards environment is currently moving towards performance-based standards, and there exists scope for such development within the relevant Australian standards, which will enable utilisation of new, 'greener' construction materials in civil engineering projects.

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BIOGRAPHY

Dr Peter Duxson BA BE (Hons), Ph D, is Business Development Manager of Zeobond Pty Ltd, a Melbourne company which has commercialised alkali-activated concrete in domestic and international markets. He is a member of the RILEM (International Union of Laboratories and Experts in Construction Materials, Systems, and Structures) Technical Committee on Alkali-Activated Materials. Email: peter@zeobond.com

Dr John Provis BE(Hons), BSc, Ph.D, is a senior research fellow in the Department of Chemical and Biomolecular Engineering, University of Melbourne, where he manages the research programme investigating alkali-activated construction materials. He is also a working group convenor within the RILEM Technical Committee on Alkali-Activated Materials.

APPENDIX – CONTACTS FOR CONCRETE

Research

University of Melbourne

Geopolymer and Minerals Processing Group www.chemeng.unimelb.edu.au/geopolymer/

Department of Chemical & Biomolecular Engineering University of Melbourne, Parkville, Vic 3010

Centre for Sustainable Resource Processing

26 Dick Perry Avenue, Kensington WA 6151

www.geopolymers.com.au

Monash University Civil Engineering

Department of Civil Engineering

Monash University, Clayton Campus, Vic 3800

civil.eng.monash.edu.au/about/staff/sanjay

Industry Contacts

Boral GPO Box 910, Sydney NSW 2001

www.boral.com.au

TecEco

497 Main Road, Glenorchy, Tasmania 7010

www.tececo.com

Zeobond

189 Northbourne Road, Campbellfield, 3061, Victoria

Corresponding author contact information:

www.zeobond.com

Zeobond Pty Ltd

PO Box 210, Somerton, Victoria 3062, Australia Tel. +61-3-9303-7777; Fax. +61-3-9303-7644;

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