Abstract

Although concrete is an essential component of sustainable building, it also contains high embodied CO$_2$. The cement and concrete industries in New Zealand are undertaking measures to manage the carbon footprint of their products. This paper outlines the three examples of projects that Holcim (New Zealand) Ltd is involved with, aimed at understanding and reducing the embodied energy contained within its products.

- An active programme of alternative fuel and raw material (AFR) use for cement production has been in place at the company’s Westport cement plant for the last 12 years. Over this time, the use of AFR (predominantly used oil) has helped reduce specific CO$_2$ emissions from cement by more than 20%.

- Research by the company indicates that up to 50% of the CO$_2$ emissions during cement manufacturing can be reabsorbed (particularly when aged concrete is crushed for recycling). It appears that recarbonation of crushed concrete in New Zealand is occurring at a similar rate of CO$_2$ uptake in demolition concrete as found by previous Nordic research. Further work is continuing to determine optimal conditions for uptake, and the timeframes over which this can be expected to occur for typical New Zealand concrete.

- An engineering feasibility study is currently being carried out on the use of algae to capture CO$_2$ and use as a fuel for cement manufacturing in New Zealand. This project has the potential to close the loop on CO$_2$ production from cement manufacturing and significantly reduce the embodied CO$_2$ of concrete products.

The cement and concrete manufacturing industries are confident in their ability to continue to supply sustainable products for New Zealand infrastructure.

Key Words: Portland cement, concrete, Holcim, embodied CO$_2$, durability, thermal comfort, Life Cycle Assessment (LCA), Alternative Fuels and Raw Materials (AFR), recarbonation, bioreactor, algae.
Introduction

Holcim Limited is one of the world’s leading suppliers of cement, aggregates, concrete and construction related services. Based in Switzerland, the Holcim Group is now a global player with a market presence in 70 countries on all continents and employing over 90,000 people. Holcim Limited is a founding member of the World Business Council for Sustainable Development, has a partnership with the World Conservation Union (IUCN) and operates the Holcim Foundation for Sustainable Construction. The company’s vision is to “provide foundations for society’s future”. Holcim (New Zealand) Ltd is a wholly owned subsidiary of Holcim Limited and is involved with the manufacturing of cement, lime, aggregates and ready mix concrete at 35 sites throughout New Zealand.

Concrete is one of the most widely used construction materials, comprising significant proportions of the built environment in many developed countries. In its simplest form, concrete is a mixture of sand and gravel (or crushed stone) and a Portland cement paste. Portland cement (cement) typically comprises 11% of the concrete mix by volume. Hydration of cement with water hardens to bind the aggregates into a rock-like material. Modern mix designs are used to ensure a range of concrete is produced to meet specific construction requirements.

Concrete is considered to be energy-efficient and eco-friendly building material, particularly when a life cycle assessment approach (LCA) is taken. The attributes of concrete as a sustainable building material include its durability and longevity, thermal comfort, fire resistance, security, ability to recycle, acoustic properties and the potential to absorb carbon dioxide (CO$_2$).

During the cement manufacturing process, carbon dioxide is produced both as a result of burning fossil fuels and as a result of the calcination process (where CaCO$_3$ is converted to CaO). Generation of CO$_2$ within the kiln is defined by the following two reactions:

\[
\text{Carbon (fuel) + O}_2 = \text{CO}_2 + \Delta H \\
\text{CaCO}_3 \text{ (usually limestone) + } \Delta H = \text{CaO} + \text{CO}_2
\]

Because of this dual generation of CO$_2$, cement and concrete are often also described as energy intensive materials with high embodied CO$_2$. The author calculates that one tonne of concrete contains approximately 102kg of embodied CO$_2$, which is predominantly a result of the cement manufacturing process. As a global industry, cement production is estimated to contribute 3.8% of all anthropogenic greenhouse gas emissions globally (Baumert et al, 2005).

In New Zealand, the cement and concrete industry is well aware of its CO$_2$ contribution and also of New Zealand’s commitments under the Kyoto Protocol. Additionally, the continued focus on green building materials, environmental labelling and plans for an emissions trading scheme have highlighted to the industry the need to understand and reduce impacts on climate change. As a result, over recent time there has been a significant drive towards sustainable practices in the sector. Production efficiencies have been introduced, cement additives and alternative fuels have been used and new technologies planned to ensure the industry understands and minimises its carbon footprint. However, the question remains as to whether these initiatives are enough to ensure that concrete can continue to be considered a sustainable product.
Alternative Fuels and Raw Materials (AFR)

Holcim operates the Used Oil Collection Programme (UORP) to collect and supply used oil to its cement plant in Westport. The programme was established in 1996 in response to a growing environmental concern that used oil was not being recovered and used responsibly in New Zealand (MfE, 2001).

Funded by Holcim, Shell, BP, Caltex, and D R Britton Ltd (Valvoline), and in partnership with the Ministry for the Environment, the UORP is designed to give oil producers, users and regulators the assurance that their used oil is being collected, transported and used or disposed of in an environmentally responsible way. Used oil is collected from hundreds of enterprises which range from oil producers, major industrial oil consumers, to small garages and workshops. Business systems are employed aimed at meeting rigorous quality standards and high levels of environmental compliance. At the cement plant, the used oil is co-processed with coal as a fuel for the production of cement.

Figure 1 shows the quantities of thermal energy provided by coal and used oil at Westport Works and the downward trend in the amount of CO\(_2\) (kg) produced per tonne of cement.

The downward trend in the amount of CO\(_2\) (kg) produced per tonne of cement (Figure 1) is assisted by two main factors: an improvement in the thermal energy efficiency of the Westport plant and the gradual increase in the quantity of used oil co-processed. During 2006, used oil provided 18.0% of total process heat for the kilns, and reduced CO\(_2\) production by 3.1% compared to coal-firing only. Co-processing of AFR (predominately used oil) has significantly helped reduce the emissions of CO\(_2\) at Westport Works when compared with operation if only coal had been used for process heat. The combustion of used oil produces approximately 74 kg CO\(_2\) per GJ of (net) thermal energy gained, while for coal the figure is 89 kg CO\(_2\) per GJ. Thus there is a reduction in CO\(_2\) production of about 17% for used oil compared with coal per unit of useful thermal energy gained.
Recent developments in the use of alternative raw materials such as millscale, ironsand tailings and waste mussel shell from aquaculture have further improved efficiencies and reduced energy use. Holcim is continuing to develop its programme of AFR and hopes to further reduce CO₂ as a result.

**Uptake of CO₂ by Concrete**

Of significance to the sustainability discussion is the ability of cement-based materials (typically concrete) to re-absorb / uptake CO₂ throughout their life. Exposed to air, these materials will absorb CO₂ over time in a process termed recarbonation. Recarbonation is likely to occur during the service life of concrete, but more importantly will occur rapidly with demolition and reprocessing. The ability of concrete to reabsorb some, or all, of this carbon dioxide has important implications for the cement and concrete industry.

The primary recarbonation reaction in concrete is given by:

\[ \text{Ca(OH)}_2 + \text{CO}_2 = \text{CaCO}_3 + \text{H}_2\text{O} \]

(Calcium silicate hydrates also contribute to the recarbonation of concrete to a lesser extent).

Research was conducted by Holcim in order to confirm whether recarbonation of concrete was a measurable phenomenon in a New Zealand context. Twenty samples of demolition and crushed concrete were collected from the Christchurch and Auckland areas late in 2007 and early in 2008. The samples were from buildings and structures dating between 0 and 84 years. The sampling was representative of the demolition materials and crushed stockpiles. Once collected, all samples where sealed in plastic until they were relocated to the laboratory for testing.

A carbonate titration method (Holcim, 2004) was adopted as the main test method for the study. A pulverized powder was digested in hydrochloric acid and heated, before being titrated with sodium hydroxide and phenolphthalein indicator. The amount of carbonate present was determined by measuring the volume of sodium hydroxide used for this reaction to occur. An additional test was undertaken on a qualitative basis to corroborate the results of the main test - where residual whole concrete samples were tested, using a (‘colour change’) phenolphthalein method (Anstice et al., 2005).

The titration results of the samples are shown in Table 1. Of the samples collected, 16 were from homogenous sources and were age verified. With the exception of the sample from the steps of the Christchurch Women’s Hospital (later found to be mortar), the titration results showed a clear trend of increasing carbonate concentration with age (Figure 2). The relationship indicates that carbonation occurs quickly initially and then slows over time. Taking a line of best fit for the valid results, a relationship between carbonation and concrete age was established to be \( C \) (\%) = 3.9 \( T^{0.33} \) (where \( C \) is percentage of carbonate measured, and \( T \) is concrete age, in years). Although the limited number of samples used in the study may not have a high degree of statistical significance, the results from the phenolphthalein indicator tests indicate a good degree of correlation with the titration tests and the trend is likely to be valid.
The tests determined the extent of recarbonation and the relationship that exists between carbon dioxide absorbed and age of the concrete. Recarbonation of New Zealand concrete appears to be occurring at a similar rate to that noted in previous Nordic research (Pade and Guimaraes, 2007).

Interestingly, the Norwegian Building Research Institute (BYGGFORSK) have examined carbonation of varying sizes of concrete under laboratory in some detail (www.danishtechnology.dk/building/14460.2). As part of their research, it was found that 60 – 80% of the CO₂ released during calcination has the potential to be chemically reabsorbed by concrete mixtures with a grain size of 1 – 8 mm, within 20 – 35 days of exposure. They also found that coarser aggregate samples carbonated at a significantly slower rate.

The results from the New Zealand samples indicate that most of the carbon dioxide emissions from calcination of limestone during cement manufacture also appear to reabsorbed (specifically when aged concrete is crushed for recycling). Further research is underway to determine optimal conditions for carbon dioxide sequestration, and the timeframes over which this can be expected to occur for New Zealand concrete. It is concluded that with the wider use of crushed recycled concrete, the effects of recarbonation should be considered in any rigorous life cycle assessment of the sustainability of concrete structures.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Age (yrs)</th>
<th>Carbonation %</th>
<th>State</th>
</tr>
</thead>
<tbody>
<tr>
<td>Chch Women’s Hospital (step)</td>
<td>57</td>
<td>24.79</td>
<td>demolition</td>
</tr>
<tr>
<td>Wallace Block</td>
<td>84</td>
<td>20.75</td>
<td>Crushed</td>
</tr>
<tr>
<td>Canterbury Saleyards</td>
<td>60</td>
<td>17.97</td>
<td>Demolition</td>
</tr>
<tr>
<td>Glassworks</td>
<td>50</td>
<td>15.27</td>
<td>Demolition</td>
</tr>
<tr>
<td>Chch Women’s Hospital (plaque)</td>
<td>57</td>
<td>14.88</td>
<td>Demolition</td>
</tr>
<tr>
<td>Chch Women’s Hospital (slab)</td>
<td>57</td>
<td>13.28</td>
<td>Demolition</td>
</tr>
<tr>
<td>Turners &amp; Growers</td>
<td>50</td>
<td>12.41</td>
<td>Demolition</td>
</tr>
<tr>
<td>Blenheim Rd Overpass</td>
<td>43</td>
<td>12.17</td>
<td>Demolition</td>
</tr>
<tr>
<td>MPL 40mm</td>
<td>Mixed</td>
<td>12.14</td>
<td>Crushed</td>
</tr>
<tr>
<td>MPL 20mm</td>
<td>Mixed</td>
<td>8.77</td>
<td>Crushed</td>
</tr>
<tr>
<td>Concrete Recyclers 65mm</td>
<td>15</td>
<td>8.39</td>
<td>Crushed</td>
</tr>
<tr>
<td>Concrete Recyclers Slab</td>
<td>15</td>
<td>8.22</td>
<td>Demolition</td>
</tr>
<tr>
<td>Fulton Hogan 65mm</td>
<td>Mixed</td>
<td>6.91</td>
<td>Crushed</td>
</tr>
<tr>
<td>Fulton Hogan 40mm</td>
<td>Mixed</td>
<td>6.12</td>
<td>Crushed</td>
</tr>
<tr>
<td>Stevensons Masonry Block 2yrs</td>
<td>2</td>
<td>6.07</td>
<td>Block</td>
</tr>
<tr>
<td>RM Core Sample 27/11</td>
<td>0.25</td>
<td>4.82</td>
<td>Core</td>
</tr>
<tr>
<td>Allied</td>
<td>0.25</td>
<td>3.59</td>
<td>Core</td>
</tr>
<tr>
<td>Stevensons Masonry Block 2Mths</td>
<td>0.17</td>
<td>3.14</td>
<td>Block</td>
</tr>
<tr>
<td>RM Core Sample 22/11</td>
<td>0.25</td>
<td>3.34</td>
<td>Core</td>
</tr>
<tr>
<td>Bombay Core Sample</td>
<td>0.1</td>
<td>0.63</td>
<td>Core</td>
</tr>
</tbody>
</table>
Holcim’s most recent research project, in combination with students from the University of Canterbury, involves a conceptual design to capture the CO$_2$ gas from cement production and its use to grow algae in a pond or bioreactor. The algae which might in turn be used as a source of fuel for the cement kiln. Various species of micro-algae have been proven to be successful in producing biofuels (Murakami and Ikenouchi, 1997).

Algae have a high productivity with comparatively low water use and are dramatically stimulated by the presence of low levels of CO$_2$. Algae absorb CO$_2$ and convert it to biomass, through rapid growth. The potential for the capture of CO$_2$ from fossil-fuel powered industry by algae has been described by previous research (eg. Nakamura et al., 2001).

The objective of the Holcim research is to prepare a conceptual design to enable capture of CO$_2$ from Holcim’s Westport Cement Kiln and deliver it to an algae pond system. The algae product could then be used as a source of fuel for the kiln. There is evidence that the levels of nitrogen oxides (NOx) and and sulphur oxides (SOx) present in flue gas emission may be used as nutrients by microalgae and that this could simplify flue gas scrubbing procedures (Nakamura et al., 2001). Some pond and reactor designs include solar concentrators and fibre optic delivery of light to different parts of the pond or bioreactor, and show very high productivity.

The design elements of the project to date have involved:
- Capture of the emissions from the kiln
- Delivery of CO$_2$ to the algae raceway pond
- Algae harvesting system
- Utilisation of algae biofuels for cement production
If successfully implemented, the proposed CO\(_2\) capture and algae system would provide a sustainable fuel source for Holcim’s Westport Plant, in addition to reducing the CO\(_2\) emissions resulting from cement manufacture. At the time of writing this paper, a ‘raceway’ pond system is the preferred design concept being developed to meet Holcim’s specific requirements. The initial aim of the project is to design a system capable of reducing CO\(_2\) gas emissions from Holcim’s cement kilns by 10% through the use of algae.

**Conclusions**

Holcim (New Zealand) Ltd is currently involved with several projects which aim to understand and minimise CO\(_2\) emissions in the short, medium and long-term.

- Current on-going co-processing of used oil has already significantly reduced the emissions of CO\(_2\) at the Westport Cement Plant. Other alternative fuels and materials are also being trialled and additional reductions are likely.

- Recarbonation of concrete in a New Zealand context has been experimentally tested by Holcim and has been determined to be a measurable, even significant, phenomenon across a range of concrete types, ages and exposure conditions. While the extent of recarbonation is affected by environmental and concrete composition parameters, the post-service demolition and crushing of concrete is considered to be an extremely significant factor in determining CO\(_2\) uptake. The recarbonation of concrete in New Zealand represents an important process which should be considered when discussing the embodied CO\(_2\) of cement and concrete. With the wider use of crushed recycled concrete, the effects of recarbonation must be considered in any rigorous life cycle assessment of the sustainability of concrete structures.

- Continuing research regarding the use of algae to capture stack gases has the potential to effectively close the loop on CO\(_2\) generation from cement manufacturing.

The inherent properties of concrete for a range of applications along with efficiencies and innovations during cement manufacturing, should help to ensure that concrete continues to be considered as a sustainable building material well into the future. Holcim (New Zealand) Ltd will continue its work on projects that will help the understanding and reduction of CO\(_2\) associated with it’s products which contribute towards sustainable solutions in New Zealand’s built environment.
References


