Optimal Low Carbon Cement Production Cost via Co-Processing and Carbon Capture and Storage

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Cement production is recorded to have released about 5% of current global man-made CO₂ emissions. They came from the burning of fossil fuel in kiln, electricity usage from grinding of raw and finished materials, and from the calcination of main raw material, limestone to produce clinker. This paper objective is to find the best minimized solution of cement production cost while reducing CO₂ emission and without compromising the quality of cement product. This is achieved by developing mathematical optimisation model that will be executed using General Algebraic Modelling System (GAMS). Some of data used in this research will be costs to install the technologies, costs of raw materials and fuels, properties of raw materials, CO₂ emission improvements from CO₂ reduction technologies and others. CO₂ reduction technologies for cement industry considered in this study are co-firing (co-process) and carbon capture and storage. This paper also discusses the best combination of energy efficient technologies to meet the CO₂ reduction target and product specification.

1. Introduction

Global warming causes by the greenhouse effect have been on an increase motion from past decades paralleling the increase of industrial activities globally. CO₂ is emitted from both manufacturing process and also from the chemical transformation of the raw materials in cement industry. As clinker production produces 90% of cement plant carbon emission; with 40% from fuels burning and 50% from chemical reaction around cement kiln (Benhelal et al., 2013), measures to reduce carbon emissions should be focused around it. One of the most common mitigation methods is co-processing. Co-processing aims to exchange carbon-based thermal sources with greener thermal sources (less carbon emissions), such as natural gas, biomass or biogenic fuels.

Wennersten et al. (2015) predict that the development of alternative fuels is still marginal in the future as opposed to fossil fuels and discussed the possibility of adapting carbon capture and storage (CCS): a technology that comprises of three stages: carbon capture, transport and storage. One of the more prominent stages consists of carbon capture stage. Carbon can be captured from products or gas streams in three ways: post combustion, oxy-fuel, and pre combustion. From an economical point of view, implementation of CCS is limited due to the addition of capital investment while environmentally; CCS is highly favourable as they are capable in reducing up to 74% CO₂ emission (Barker et al., 2009). Other possibility was suggested by Gul et al. (2014) where locally available mineral material (LAMM) that possess cementitious properties was added to cement clinker to reduce energy consumption. The result shows that replacement of 15% clinker with LAMM reduces the thermal energy consumption for clinkerisation and simultaneously the CO₂ emissions.

An optimisation model was proposed by Kookos et al. (2011) to find the minimized cement manufacturing costs with the implementation of co-processing. The study, however, only discusses the possibility of co-processing in cement plant without exploring other mitigation methods. To improve this, additional mitigation method; CCS, is included in the model to study the effects of implementing one or more mitigation methods on various parameters such as production costs and carbon reduction.
2. Model Formulation

2.1 Objective function

The objective is to find the minimize solution of production cost in €/t clinker that satisfy a CO\(_2\) reduction target while maintaining product quality. The cost functions in this problem include capital cost of running a fully functioned cement plant, cost of raw materials, cost of fuels, and the retrofitting cost associated with co-processing, and CCS. The objective function for this is given by

\[
Z_{\text{min}} = FCI + \sum_{j \in \text{Raw materials}} C_j m_j + \sum_{k \in \text{Fossil}} C_k m_k + \sum_{l \in \text{Nonfossil}} C_l m_l + \sum_{c \in \text{CCS}} (FCI, X_c + VC_c m_l) + \sum_{c \in \text{CCS}} (FCI, X_c + VC_c) \tag{1}
\]

where FCI is the fixed capital cost in €/t clinker. The model involves the product of raw materials j costs; \(C_j\) in €/kg of raw materials j, with the mass of raw materials \(m_j\) in kg/t clinker produced. The product produces the cost of raw materials j used in producing t clinker. To find the cost of fuels used, there are two types of fuels involved in cement production which are fossil fuels \(k\) and alternative fuels \(l\). \(C_k\) and \(C_l\) in the model represents cost of fuels \(k\) and cost of fuels \(l\) in €/kg of fuels while \(m_k\) and \(m_l\) represents mass of fuels \(k\) and mass of fuels \(l\) in kg/t clinker produced. The model also includes the retrofitting cost. Retrofitting cost in this study includes the cost in handling, storage, installation, facilities for alternative fuels \(l\), and retrofitting costs for new CCS technologies \(c\) facilities. FCI and FCI\(_c\) is the fixed capital investment for non-fossil fuels \(l\) in €/t clinker and CCS technologies \(c\) in €/t clinker produced while VC\(_c\) and VC\(_C_c\) is the variable cost for non-fossil fuels \(l\) in €/kg of fuels and CCS technologies \(c\) in €/t clinker. Binary variables \(X_k\) and \(X_c\) are introduced for selection of fuels and CCS technologies.

2.2 Constraints

The objective function is subjected to few constraints.

Oxides, alkalis, sulphur and heavy metals produced in kiln came from raw materials and ash from the burning of fuels. They enter the system through raw meals and ash from fuels while sulphurs come through raw meals and the fuels themselves. The constraint is formulated as:

\[
\text{clinker production} = \sum_{o \in \text{Oxides}} m_o + \sum_{a \in \text{Alkalides}} m_a + \sum_{s \in \text{Sulphurs}} m_s + \sum_{h \in \text{Heavy Metals}} m_h \tag{2}
\]

where \(m_o\), \(m_a\), \(m_s\), and \(m_h\) represents the total mass of oxides, total mass of alkalis, total mass of sulphurs and total mass of heavy metals kg/t clinker.

Oxides in clinker produced are subjected to:

\[
m_o = \sum_{j \in \text{Raw materials}} \omega_{o,j} m_j + \sum_{k \in \text{Fossil}} \omega_{o,k} m_k + \sum_{l \in \text{Nonfossil}} \omega_{o,l} m_l \tag{3}
\]

where \(\omega_{o,j}\) is the mass fraction of oxides \(o\) in raw materials \(j\), \(\omega_{o,k}\) is the mass fraction of oxides \(o\) in fossil fuels’ ash \(k\) and \(\omega_{o,l}\) is the mass fraction of oxides \(o\) in alternative fuels’ ash \(l\). Mass of alkalis and heavy metals are also subjected to the same formula as in Eq.(3). Mass fractions are in wt.\%.

Sulphurs in clinker produced are subjected to the difference between sulphurs produced from raw materials, and fuels with the amount of sulphurs released during combustion as shown below:

\[
m_s = \sum_{j \in \text{Raw materials}} \omega_{s,j} m_j + \sum_{k \in \text{Fossil}} \omega_{s,k} \times \frac{80}{32} m_k + \sum_{l \in \text{Nonfossil}} \omega_{s,l} \times \frac{80}{32} m_l - V_{fg} \cdot \text{Conc}_{SO_3} \tag{4}
\]

where \(\omega_{s,j}\) is the mass fraction of sulphurs \(s\) in raw materials \(j\), \(\omega_{s,k}\) is the mass fraction of sulphurs \(s\) in fossil fuels \(k\) and \(\omega_{s,l}\) is the mass fraction of sulphurs \(s\) in non-fossil fuels \(l\). The mass fractions of the sulphurs in fuels are calculated by expressing the S wt.% in fuels \(k\) and \(l\) to SO\(_3\) wt.% by introducing 80 kg SO\(_3\)/mol divided by 32 kg S/mol. Amount of SO\(_3\) in flue gas is calculated by the product of flue gas volumetric flow rate, \(V_{fg}\) in Nm\(^3\)/t clinker with concentration of SO\(_3\) in flue gas in kg/Nm\(^3\).

Volumetric flowrate of flue gas is calculated by assuming an ideal gas law under normal condition:

\[
V_{fg} = 22.414 \sum_{f \in \text{Flue Gases}} \frac{m_f}{m_{w_f}} \tag{5}
\]

where \(m_{w_f}\) is the mass of flue gas \(f\) in kg, \(m_{w_f}\) is the molecular weight of flue gas \(f\) in kg/kmol. 22.414 is the molar volume of an ideal gas under normal condition, in Nm\(^3\)/kmol.
Flue gases emitted from cement plant are NO\textsubscript{2}, CO\textsubscript{2}, and SO\textsubscript{3}. The flue gases are formed via fuels combustion (CO\textsubscript{2}, NO\textsubscript{2}, and SO\textsubscript{3}), and chemical reaction from clinkerisation (CO\textsubscript{2}). Assuming that air fed for combustion consists of 76.8 % N\textsubscript{2} and 23.2 % O\textsubscript{2}, with O\textsubscript{2} released is controlled at a level of 10 %, and complete combustion is achieved, constraints gathered are as follows:

\[ m_{N_2} = 76.8\% \times m_{air} \quad (6) \]

\[ m_{O_2} = (23.2\% \times m_{air}) - \left( \sum_{k \in \text{Fossil}} S_l m_k + \sum_{l \in \text{Nonfossil}} S_l m_l \right) \quad (7) \]

\[ m_{O_2} = V_{fr} \left( \frac{32}{22.414} \right)(10\%) \quad (8) \]

where \( m_{air} \) represents mass of air fed in kg/t clinker. O\textsubscript{2} is calculated as the difference between O\textsubscript{2} in fed air in kg/t clinker with O\textsubscript{2} used for complete combustion in kg/t clinker. \( S_l \) and \( S_t \) are the stoichiometric O\textsubscript{2} required for complete combustion of fuels \( k \) and \( l \). \( S_t \) and \( S_t \) are calculated as per Green and Perry (2008).

As stated before, CO\textsubscript{2} emissions are contributed by the clinkerisation process and combustion of fuels. CO\textsubscript{2} emissions from fuels are a combination of CO\textsubscript{2} emissions from fossil fuels and CO\textsubscript{2} emissions from non-fossil fuels. Mass of CO\textsubscript{2} emitted from clinkerisation \( (m_{cl}) \) and combustion \( (m_{cm}) \) are formulated as:

\[ m_{cl} = \frac{44}{55} \sum_{j \in \text{Raw materials}} \omega_{\text{CaO},j} m_j + \frac{44}{40} \sum_{j \in \text{Raw materials}} \omega_{\text{MgO},j} m_j \quad (9) \]

\[ m_{cm} = \sum_{k \in \text{CEF}} \alpha_k + \sum_{l \in \text{Nonfossil}} \alpha_l \quad (10) \]

\[ CO_{2_{\text{total}}} = m_{cl} + m_{cm} \quad (11) \]

where \( \omega_{\text{CaO},j} \) is the mass fraction of CaO in raw materials \( j \) and \( \omega_{\text{MgO},j} \) is the mass fraction of MgO in raw materials \( j \). Both 44/55 and 44/40 are introduced since CaO and MgO are used as the basis of the calculation (the same principal used to calculate S in SO\textsubscript{3} in Eq(4)). CEF\( k \) is the carbon emission factor of fossil fuels \( k \) and CEF\( l \) is the carbon emission factor of non-fossil fuels \( l \). \( \alpha_k \) and \( \alpha_l \) are the product between \( m_k \) with binary variable \( X_k \), and \( m_l \) with binary variable \( X_l \). Product of two continuous variables resulted in nonlinear models. In an effort to linearised nonlinear models, each continuous variable \( (\alpha_k \) and \( \alpha_l) \) are then subjected to constraints proposed by Adams et al. (2004). Summation of Eq(9) and Eq(10) generates total CO\textsubscript{2} emissions (Eq(11)), in kg/t clinker.

Heat consumed in kilns is supplied by both fuels where NCV\( k \) is the net calorific value of fossil fuels \( k \) in GJ/kg of fuels and NCV\( l \) is the net calorific value of non-fossil fuels \( l \) in GJ/kg of fuels. TED represents the thermal energy demand in GJ/t clinker. This gives:

\[ TED = \sum_{k \in \text{CEF}} \text{NCV}_k m_k + \sum_{l \in \text{Nonfossil}} \text{NCV}_l m_l \quad (12) \]

The use of alternative fuels is subjected to permits; thermal substitution rate (TSR) as shown:

\[ \sum_{l \in \text{Nonfossil}} \text{NCV}_l m_l \leq (\text{TSR\%}) TED \quad (13) \]

Based on clinker analysis, there are assumed to be 4 major phases (p) in clinker; C\textsubscript{3}S, C\textsubscript{2}S, C\textsubscript{3}A and C\textsubscript{4}AF formed by oxides from the raw materials:
\[ M_{pL} \leq \sum_{j \in \text{Raw materials}} \text{Bogue} \cdot m_{o,j} + \sum_{k \in \text{Fossil}} \text{Bogue} \cdot m_{o,k} + \sum_{l \in \text{Nonfossil}} \text{Bogue} \cdot m_{o,l} \leq M_{pU} \]  \hspace{1cm} (14)

where \( M_{pL} \) in kg/t clinker represent the lower limits for clinker phases \( \text{C}_3\text{S}, \text{C}_2\text{S}, \text{C}_3\text{A} \) and \( \text{C}_4\text{AF} \) while \( M_{pU} \), in kg/t clinker represent the upper limits. \( m_{o,j}, m_{o,k}, \) and \( m_{o,l} \) are the mass of oxides in raw materials \( j \), fossil fuels’ ash \( k \) and non-fossil fuels’ ash \( l \). Bogue value is obtained from Kookos et al. (2011).

Environmental constraint is formulated for total carbon emissions to not be more than proposed carbon emission reduction. This is achieved by:

\[
\text{CO}_{2\text{total}} - \sum_{c \in \text{CCS}} \beta_c \alpha_c \leq (1 - \%\text{CO}_2) \text{CO}_{2\text{target}}
\]

\hspace{1cm} (15)

where \( \text{CO}_{2\text{total}} \) is the amount of \( \text{CO}_2 \) emitted gained from Eq(11). \( \%\text{CO}_2 \) is the emission reduction target and \( \text{CO}_{2\text{target}} \) is current \( \text{CO}_2 \) emission in kg/t clinker. \( \beta_c \) is the percent reduction when CCS \( c \) is implemented. \( \alpha_c \) is the linearization variable; product of binary variable \( X_c \) with \( \text{CO}_{2\text{total}} \) and subjected to constraints proposed by Adams et al., 2004.

Fuels usage must not exceed the availability of the fuels.

\[ m \leq AX \]  \hspace{1cm} (16)

where \( A \) is the availability of fuels in kg/y.

The number of fuels mixed must not exceed the maximum amount of fuels that can be mixed.

\[ X_k + X_j \leq \text{max fuels mixture} \]  \hspace{1cm} (17)

\( X_k \) and \( X_j \) are the binary variables for the use of fuels.

The number of CCS installed must not exceed 1.

\[ X_c \leq 1 \]  \hspace{1cm} (18)

\( X_c \) is the binary variable for the use of CCS.

All the binaries are subjected to:

\[
X \begin{cases} 1 & \text{if technology is used} \\ 0 & \text{otherwise} \end{cases}
\]

\hspace{1cm} (19)

2.3 Case study

Cement plant with an annual capacity of 1 Mt Ordinary Portland Cement/y plant with 5-stage pre-heater pre-calcerin kiln system. Raw materials options are limestone, clay, sand, iron source while fuels options are coal, petroleum coke (PC), refuse derived fuel (RDF), sewage sludge (SS), tire derived fuel (TDF), meat bone meal (MBM). CCS technologies options are post combustion and oxy-fuel. For the base plant, no alternative fuels are used. TSR value of 40 \% is used. Data are gathered from Kookos et al. (2011) for chemical analysis of raw materials and fuels and fuels price, U.S. Geological Survey (2012) for clay, sand and iron source prices, Willett (2011) for limestone price, Moya et al. (2010) for plant FCI value, and Barker et al. (2009) for CCS data. The models are executed in GAMS by changing the reduction target. The optimisation stops when infeasible result is achieved indicating maximum possible \( \text{CO}_2 \) reduction achieved.

3. Result and Discussion

Results for this study are shown by Table 1 and Table 2. Table 1 includes the base case results (\%\text{CO}_2 is 0 \% and TSR 0 \%) and later optimisation right before the selection of CCS technologies while Table 2 shows the results of optimisation when CCS technologies selection is involved.

Table 1 shows that, at base case; a cement plant with no alternative fuels nor CCS technologies installed, the production cost is € 131.266 /t clinker. The \( \text{CO}_2 \) emitted is 859.984 kg \( \text{CO}_2 \)/t clinker, with 325 kg \( \text{CO}_2 \)/t clinker and 534.984 kg \( \text{CO}_2 \)/t clinker generated from fuels combustion and clinkerisation. Since no CCS is installed, amount of \( \text{CO}_2 \) produced from cement kiln is the same with \( \text{CO}_2 \) released to the atmosphere.
Installation of alternative fuels facilities is applied after TSR amount is set reducing the CO\textsubscript{2} emission by 3.1 \% and simultaneously increasing the production cost; € 129.389 /t clinker. As the reduction target is increase; 4 \%; 5 \%; 5.9702 \%, the minimized costs also increase; € 130.03 /t clinker, € 131.311 /t clinker, and € 136.396 /t clinker. The cost increases because the prior coal used to replace the higher carbon content PC increase as the CO\textsubscript{2} reduction target increase. Large cost gap between 5 \% and 5.9702 \% (€ 131.311 /t clinker and € 136.396 /t clinker) reduction target as shown in Table 2 is because post combustion CCS is chosen to be installed at 5.9702 \%. It can be seen that post combustion CCS is chosen when the amount of fuels needed for cement kiln has reached its maximum; 59.091 kg PC/t clinker (maximum CO\textsubscript{2} reduction that can be achieved from co-processing). The cost to achieve CO\textsubscript{2} reduction between 5.9702 \% and 62.1862 \% did not change since the amount of CO\textsubscript{2} needed to be reduced is always fulfilled by post combustion CCS capturing abilities. Maximum amount of CO\textsubscript{2} that can be captured by post combustion CCS is achieved around 62.1862 \%, since from 62.1862 \%, 62.2 \%, and 63 \%, manufacturing costs changes; € 136.4 /t clinker, € 136.425 /t clinker, and € 138.78 /t clinker, where amounts of coal used increase as the reduction increase. At 64 \% CO\textsubscript{2} reduction, maximum amount of fuels needed and limits for CO\textsubscript{2} capturing abilities by post combustion CCS is reached, making oxy-fuel CCS chosen as one of the mitigation strategy. The cost is higher; € 161.852 /t clinker, since higher installation cost is needed for oxy-fuel CCS. Highest amount of CO\textsubscript{2} that can be reduced in this study is 75 \%, at € 162.43 /t clinker.

4. Conclusions
In conclusion, highest emission reduction that can be achieved is 75 \% with cost of € 162.43 /t clinker. The cost is higher than base case since in order to maintain the quality of clinker produced while submitting to carbon reduction target, various facilities have to be installed. This study can be further explored by considering the effect of installing CCS to the feeding materials. In this study, the effect from CCS is only in respect to the amount CO\textsubscript{2} reduction, while in real practice, installing CCS led to the change of air fed affecting the amount of flue gases emitted from cement kiln.

Table 1: Results for base case scenario and after optimisation without CCS technologies selection

<table>
<thead>
<tr>
<th>Reduction target (%CO\textsubscript{2})</th>
<th>Only fossil</th>
<th>II</th>
<th>III</th>
<th>IV</th>
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<tbody>
<tr>
<td>TSR (%)</td>
<td>0</td>
<td>0.031</td>
<td>0.04</td>
<td>0.05</td>
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<tr>
<td>Cost (€/t clinker)</td>
<td>131.266</td>
<td>129.389</td>
<td>130.03</td>
<td>131.311</td>
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<tr>
<td>CO\textsubscript{2} produced (kg CO\textsubscript{2}/t clinker)</td>
<td>859.984</td>
<td>833.324</td>
<td>825.585</td>
<td>816.985</td>
</tr>
<tr>
<td>Fossil fuels</td>
<td>325</td>
<td>194.408</td>
<td>186.003</td>
<td>178.75</td>
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<tr>
<td>Nonfossil fuels</td>
<td>0</td>
<td>104</td>
<td>104</td>
<td>104</td>
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<tr>
<td>Clinkerization</td>
<td>534.984</td>
<td>534.916</td>
<td>535.582</td>
<td>534.235</td>
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<tr>
<td>CO\textsubscript{2} released (kg CO\textsubscript{2}/t clinker)</td>
<td>859.984</td>
<td>833.324</td>
<td>825.585</td>
<td>816.985</td>
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<tr>
<td>Raw materials used (kg/t clinker)</td>
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<td>1,486.079</td>
<td>1,486.404</td>
<td>1,484.746</td>
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<td>1,314.461</td>
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<td>0</td>
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<td>Sand</td>
<td>161.397</td>
<td>160.139</td>
<td>157.422</td>
<td>131.355</td>
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<td>Iron source</td>
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<td>11.479</td>
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<td>Fuels used (kg/t clinker)</td>
<td>98.485</td>
<td>99.931</td>
<td>102.988</td>
<td>105.625</td>
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<tr>
<td>Fossil fuels</td>
<td>98.485</td>
<td>59.306</td>
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<tr>
<td>Petroleum coke</td>
<td>98.485</td>
<td>56.938</td>
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<td>40.625</td>
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<tr>
<td>TDF</td>
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<td>40.625</td>
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Table 2: Results for optimisation with CCS technologies selection

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<tr>
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<th>VII</th>
<th>VIII</th>
<th>IX</th>
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<td>Reduction target (%CO₂)</td>
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<td>TSR (%)</td>
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<tr>
<td>Cost (£/t clinker)</td>
<td>136.396</td>
<td>136.4</td>
<td>136.425</td>
<td>138.78</td>
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<td>162.43</td>
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<td>CO₂ produced (kg CO₂/t clinker)</td>
<td>833.87</td>
<td>833.827</td>
<td>833.523</td>
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<td>833.87</td>
<td>833.87</td>
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<tr>
<td>TSR (%)</td>
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<td>0.4</td>
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<tr>
<td>CO₂ captured (kg CO₂/t clinker)</td>
<td>508.661</td>
<td>508.634</td>
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<td>CO₂ released (kg CO₂/t clinker)</td>
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<td>325.074</td>
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<td>Sand</td>
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<td>160.315</td>
<td>160.209</td>
<td>117.745</td>
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<td>157.886</td>
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<td>Fuels used (kg/t clinker)</td>
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<td>99.733</td>
<td>99.853</td>
<td>105.625</td>
<td>99.716</td>
<td>102.466</td>
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<td>59.091</td>
<td>65</td>
<td>59.091</td>
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<td>Oxy-fuel combustion</td>
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Acknowledgment

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References


