

# VOL. 45, 2015



Guest Editors: Petar Sabev Varbanov, Jiří Jaromír Klemeš, Sharifah Rafidah Wan Alwi, Jun Yow Yong, Xia Liu Copyright © 2015, AIDIC Servizi S.r.I., ISBN 978-88-95608-36-5; ISSN 2283-9216

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

# Siti A. Ishak\*, Haslenda Hashim, Zarina Ab. Muis

Process Systems Engineering Centre (PROSPECT), Faculty of Chemical Engineering, Universiti Teknologi Malaysia, 81310 Skudai, Johor, Malaysia siti.aktar@gmail.com

Cement production is recorded to have released about 5 % of current global man-made  $CO_2$  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  $CO_2$  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,  $CO_2$  emission improvements from  $CO_2$  reduction technologies and others.  $CO_2$  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  $CO_2$  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. CO<sub>2</sub> 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 carbonemissions), 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 % CO<sub>2</sub> 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 CO<sub>2</sub> 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.

DOI: 10.3303/CET1545050

#### 2. Model Formulation

#### 2.1 Objective function

The objective is to find the minimize solution of production cost in  $\in$ /t clinker that satisfy a CO<sub>2</sub> 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

$$Z_{\min} = FCI + \sum_{j \in Raw materials} C_j m_j + \sum_{k \in Fossil} C_k m_k + \sum_{l \in Nonfossil} C_l m_l + \sum_{l \in Nonfossil} (FCI_l X_l + VC_l m_l) + \sum_{c \in CCS} (FCI_c X_c + VC_c)$$
(1)

where FCI is the fixed capital cost in  $\notin$ /t clinker. The model involves the product of raw materials j costs; C<sub>j</sub> in  $\notin$ /kg of raw materials j, with the mass of raw materials j; m<sub>j</sub> 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 I. C<sub>k</sub> and C<sub>l</sub> in the model represents cost of fuels k and cost of fuels I in  $\notin$ /kg of fuels while m<sub>k</sub> and m<sub>l</sub> represents mass of fuels k and mass of fuels I in kg/t clinker produced. The model also includes the retrofitting cost. Retrofitting costs for new CCS technologies c facilities. FCl<sub>l</sub> and FCl<sub>c</sub> is the fixed capital investment for non-fossil fuels I in  $\notin$ /t clinker and CCS technologies c in  $\notin$ /t clinker produced while VC<sub>l</sub> and VC<sub>c</sub> is the variable cost for non-fossil fuels I in  $\notin$ /kg of fuels and CCS technologies c in  $\notin$ /t clinker. Binary variables X<sub>l</sub> and X<sub>c</sub> 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:

clinker production = 
$$\sum_{o \in Oxides} m_o + \sum_{a \in Alkalides} m_a + \sum_{s \in Sulphurs} m_s + \sum_{h \in HeavyMetab} m_h$$
 (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:

$$m_{o} = \sum_{j \in Rawmaterials} \omega_{o,j} m_{j} + \sum_{k \in Fossil} \omega_{o,k} m_{k} + \sum_{l \in Nonfossil} \omega_{o,l} m_{l}$$
(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 I. 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:

$$m_{s} = \sum_{j \in Raw \, materials} \omega_{s,j} m_{j} + \sum_{k \in Fossil} \left( \omega_{s,k} \times \frac{80}{32} \right) m_{k} + \sum_{l \in Nonfossil} \left( \omega_{s,l} \times \frac{80}{32} \right) m_{l} - V_{fg} Conc_{SO_{3}}$$
(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 I. The mass fractions of the sulphurs in fuels are calculated by expressing the S wt.% in fuels k and l to SO<sub>3</sub> wt.% by introducing 80 kg SO<sub>3</sub>/mol divided by 32 kg S/mol. Amount of SO<sub>3</sub> in flue gas is calculated by the product of flue gas volumetric flow rate, V<sub>fg</sub> in Nm<sup>3</sup>/t clinker with concentration of SO<sub>3</sub> in flue gas in kg/Nm<sup>3</sup>.

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

$$V_{fg} = 22.414 \sum_{f \in Flue \ Gases} \frac{m_{fg}}{mw_{fg}}$$
(5)

where  $m_{fg}$  is the mass of flue gas  $f_g$  in kg.  $mw_{fg}$  is the molecular weight of flue gas  $f_g$  in kg/kmol. 22.414 is the molar volume of an ideal gas under normal condition, in Nm<sup>3</sup>/kmol.

296

Flue gases emitted from cement plant are NO<sub>2</sub>, CO<sub>2</sub>, and SO<sub>3</sub>. The flue gases are formed via fuels combustion (CO<sub>2</sub>, NO<sub>2</sub>, and SO<sub>3</sub>), and chemical reaction from clinkerisation (CO<sub>2</sub>). Assuming that air fed for combustion consists of 76.8 % N<sub>2</sub> and 23.2 % O<sub>2</sub>, with O<sub>2</sub> released is controlled at a level of 10 %, and complete combustion is achieved, constraints gathered are as follows:

$$m_{N_{\gamma}} = 76.8\% \times m_{air} \tag{6}$$

$$m_{O_2} = \left(23.2\% \times m_{air}\right) - \left(\sum_{k \in Fossil} St_k m_k + \sum_{l \in Nonfossil} St_l m_l\right)$$
(7)

$$m_{O_2} = V_{fg} \left(\frac{32}{22.414}\right) (10\%) \tag{8}$$

where  $m_{air}$  represents mass of air fed in kg/t clinker.  $O_2$  is calculated as the difference between  $O_2$  in fed air in kg/t clinker with  $O_2$  used for complete combustion in kg/t clinker. St<sub>k</sub> and St<sub>l</sub> are the stoichiometric  $O_2$  required for complete combustion of fuels k and l. St<sub>k</sub> and St<sub>l</sub> are calculated as per Green and Perry (2008).

As stated before,  $CO_2$  emissions are contributed by the clinkerisation process and combustion of fuels.  $CO_2$  emissions from fuels are a combination of  $CO_2$  emissions from fossil fuels and  $CO_2$  emissions from non-fossil fuels. Mass of  $CO_2$  emitted from clinkerisation (m<sub>cl</sub>) and combustion (m<sub>cm</sub>) are formulated as:

$$m_{cl} = \frac{44}{55} \sum_{j \in Raw \ materials} \omega_{CaO,j} m_j + \frac{44}{40} \sum_{j \in Raw \ materials} \omega_{MgO,j} m_j \tag{9}$$

$$m_{cm} = \sum_{k \in Fossil} CEF_k \alpha_k + \sum_{l \in Nonfossil} CEF_l \alpha_l$$
(10)

$$CO_{2_{T_{okl}}} = m_{cl} + m_{cm} \tag{11}$$

where  $\omega_{CaO,j}$  is the mass fraction of CaO in raw materials j and  $\omega_{MgO,j}$  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<sub>3</sub> in Eq(4)). CEF<sub>k</sub> is the carbon emission factor of fossil fuels k and CEF<sub>1</sub> is the carbon emission factor of non-fossil fuels I.  $\alpha_k$  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<sub>2</sub> 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:

$$TED = \sum_{k \in Fossil} NCV_k m_k + \sum_{l \in Nonfossil} NCV_l m_l$$
(12)

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

$$\sum_{l \in Nonfossil} NCV_l m_l \le (TSR\%) TED$$
(13)

Based on clinker analysis, there are assumed to be 4 major phases (p) in clinker; C<sub>3</sub>S, C<sub>2</sub>S, C<sub>3</sub>A and C<sub>4</sub>AF formed by oxides from the raw materials:

297

298

$$M_{p_{L}} \leq \sum_{j \in Raw \ materials} Bogue_{p} m_{o,j} + \sum_{k \in Fossil} Bogue_{p} m_{o,k} + \sum_{l \in Nonfossil} Bogue_{p} m_{o,l} \leq M_{p}^{U}$$
(14)

where  $M_{p_L}$ ; in kg/t clinker represent the lower limits for clinker phases C<sub>3</sub>S, C<sub>2</sub>S, C<sub>3</sub>A and C<sub>4</sub>AF while  $M_p^U$ ; 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 I. 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:

$$CO_{2_{Total}} - \sum_{c \in CCS} \beta_c \alpha_c \le (1 - \% CO_2) CO_{2_{GHG}}$$

$$\tag{15}$$

where  $CO_{2_{Total}}$  is the amount of  $CO_2$  emitted gained from Eq(11). %CO<sub>2</sub> is the emission reduction target and  $CO_{2_{GHG}}$  is current  $CO_2$  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  $CO_{2_{Total}}$  and subjected to constraints proposed by Adams et al., 2004.

Fuels usage must not exceed the availability of the fuels.

$$m \le AX \tag{16}$$

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.

$$X_k + X_l \le \max \text{ fuels mixture} \tag{17}$$

 $X_k$  and  $X_l$  are the binary variables for the use of fuels. The number of CCS installed must not exceed 1.

$$X_c \le 1 \tag{18}$$

 $X_{c}\xspace$  is the binary variable for the use of CCS. All the binaries are subjected to:

$$X \begin{cases} 1 \text{ if technology is used} \\ 0 \text{ otherwise} \end{cases}$$
(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-calciner 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 CO<sub>2</sub> 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 ( $%CO_2$  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  $\in$  131.266 /t clinker. The CO<sub>2</sub> emitted is 859.984 kg CO<sub>2</sub>/t clinker, with 325 kg CO<sub>2</sub>/t clinker and 534.984 kg CO<sub>2</sub>/t clinker generated from fuels combustion and clinkerisation. Since no CCS is installed, amount of CO<sub>2</sub> produced from cement kiln is the same with CO<sub>2</sub> released to the atmosphere.

Installation of alternative fuels facilities is applied after TSR amount is set reducing the CO<sub>2</sub> 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 pricier coals used to replace the higher carbon content PC increase as the CO₂ 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<sub>2</sub> reduction that can be achieved from co-processing). The cost to achieve CO<sub>2</sub> reduction between 5.9702 % and 62.1862 % did not change since the amount of CO2 needed to be reduced is always fulfilled by post combustion CCS capturing abilities. Maximum amount of CO2 that can be captured by post combustion CCS is achieved around 62.1862 %, since from 62.1862 %, 62.2 %, and 63 %, manufacturing costs changes;  $\in$  136.4 /t clinker,  $\in$  136.425 /t clinker, and  $\in$  138.78 /t clinker, where amounts of coal used increase as the reduction increase. At 64 % CO<sub>2</sub> reduction, maximum amount of fuels needed and limits for CO<sub>2</sub> 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<sub>2</sub> 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  $\in$  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<sub>2</sub> reduction, while in real practice, installing CCS led to the change of air fed affecting the amount of flue gases emitted from cement kiln.

	Only fossil	II		IV	
Reduction target	0	0.021	0.04	0.05	
(%CO <sub>2</sub> )	0	0.031	0.04		
TSR (%)	0	0.4	0.4	0.4	
Cost (€/t clinker)	131.266	129.389	130.03	131.311	
CO <sub>2</sub> produced	950 094	022 224	925 595	916 095	
(kg CO <sub>2</sub> /t clinker)	009.904	033.324	020.000	010.905	
Fossil fuels	325	194.408	186.003	178.75	
Nonfossil fuels	0	104	104	104	
Clinkerization	534.984	534.916	535.582	534.235	
CO <sub>2</sub> released	850 081	922 224	925 595	816 085	
(kg CO <sub>2</sub> /t clinker)	009.904	000.024	020.000	010.905	
Raw materials used	1 / 86 807	1 / 86 079	1 486 404	1 181 716	
(kg/t clinker)	1,400.007	1,400.073	1,400.404	1,-07-0	
Limestone	1,314.526	1,314.461	1,316.352	1,308.336	
Clay	0	0	0	29.492	
Sand	161.397	160.139	157.422	131.355	
Iron source	10.884	11.479	12.63	15.563	
Fuels used	09 495	00.021	102 099	105 625	
(kg/t clinker)	90.405	99.931	102.900	105.025	
Fossil fuels	98.485	59.306	62.363	65	
Coal	0	2.368	35.99	65	
Petroleum coke	98.485	56.938	26.373	0	
Non fossil fuels	0	40.625	40.625	40.625	
TDF	0	40.625	40.625	40.625	

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

Table 2: Results for optimisation with CCS technologies selection

	V	VI	VII	VIII	IX	Х
Reduction target	0 059702	0 621862	0.622	0.63	0.64	0.75
(%CO <sub>2</sub> )	0.0001.02	0.021002	0.022	0.00	0.04	0.70
TSR (%)	0.4	0.4	0.4	0.4	0.4	0.4
Cost (€/t clinker)	136.396	136.4	136.425	138.78	161.852	162.43
CO <sub>2</sub> produced	833 87	833.827	833.523	815.882	833.87	826.908
(kg CO <sub>2</sub> /t clinker)	000.01					
Fossil fuels	195	194.954	194.624	178.75	195	187.439
Nonfossil fuels	104	104	104	104	104	104
Clinkerisation	534.87	534.873	534.899	533.132	534.87	535.468
CO <sub>2</sub> captured	508.661	508.634	508.449	497.688	617.064	611.912
(kg CO <sub>2</sub> /t clinker)			0001110		0111001	0
CO <sub>2</sub> released	325 209	325 193	325 074	318 194	216 806	214 996
(kg CO <sub>2</sub> /t clinker)	020.200	020.100	020.071	010.101	210.000	2110000
Raw materials used	1.486.055	1.486.057	1.486.07	1.483.635	1.486.055	1.486.348
(kg/t clinker)	1,1001000	1,100.001	1,100.01	1,100.000	1,100.000	1,1001010
Limestone	1,314.328	1,314.338	1,314.412	1,302.801	1,314.328	1,316.029
Clay	0	0	0	46.412	0	0
Sand	160.33	160.315	160.209	117.745	160.33	157.886
Iron source	11.397	11.404	11.449	16.677	11.397	12.433
Fuels used	00 716	00 733	00 853	105 625	00 716	102 466
(kg/t clinker)	33.710	99.755	99.000	105.025	33.710	102.400
Fossil fuels	59.091	59.108	59.228	65	59.091	61.841
Coal	0	0.184	1.506	65	0	30.243
Petroleum coke	59.091	58.924	57.722	0	59.091	31.598
Non fossil fuels	40.625	40.625	40.625	40.625	40.625	40.625
TDF	40.625	40.625	40.625	40.625	40.625	40.625
Post combustion	0	0	0	0	•	•
Oxy-fuel combustion	٠	•	•	•	0	0

#### Acknowledgment

Special thanks to Universiti Teknologi Malaysia (UTM) and the Ministry of Higher Education of Malaysia. This research is funded by GUP research grant Vot number Q.J130000.2544.06H73.

#### References

- Adams W.P., Forrester R.J., Glover F.W., 2004, Comparisons and enhancement strategies for linearizing mixed 0-1 quadratic programs, Discrete Optimization, 1(2), 99 120.
- Barker D.J., Turner S.A., Napier-Moore P.A., Clark M., Davison J.E., 2009, CO<sub>2</sub> Capture in the Cement Industry, Energy Procedia, 1(1), 87 94.
- Benhelal E., Zahedi G., Shamsaei E., Bahadori A., 2013, Global strategies and potentials to curb CO<sub>2</sub> emissions in cement industry, Journal of Cleaner Production, 51, 142 161.
- Gul S., Saeed F., Amin N.U., 2014, Conversion of Low Cost Mineral Material to Pozzolana and its Impact on Cement Parameters and Cost of Production, Chemical Engineering Transactions, 39, 1225 – 1230.
- Kookos I.K., Pontikes Y., Angelopoulos G.N., Lyberatos G., 2011, Classical and alternative fuel mix optimization in cement production using mathematical programming, Fuel, 90(3), 1277 1284.
- Moya J.A., Pardo N., Mercier A., 2010, Energy Efficiency and CO<sub>2</sub> Emissions: Prospective Scenarios for the Cement Industry, Publications Office of the European Union, Luxembourg.

Green D.W., Perry R.H., 2008, Perry's Chemical Engineers' Handbook, 8th ed, McGraw Hill, NY, USA.

- U.S. Geological Survey, 2012, Mineral commodity summaries 2012, U.S. Department of the Interior, US Geological Survey, USA.
- Wennersten R., Sun Q., Li H., 2015, The future potential for Carbon Capture and Storage in climate change mitigation – an overview from perspectives of technology, economy and risk, Journal of Cleaner Production, 103, 724 – 736.
- Willett J.C., 2011, 2010 Minerals Yearbook: STONE, CRUSHED, US Department of the Interior, US Geological Survey, USA.