CHEMICAL ENGINEERING TRANSACTIONS

VOL. 35, 2013

A publication of
AIDIC
The Italian Association of Chemical Engineering
www.aidic.it/cet

Guest Editors: Petar Varbanov, Jiří Klemeš, Panos Seferlis, Athanasios I. Papadopoulos, Spyros Voutetakis

ISBN 978-88-95608-26-6; **ISSN** 1974-9791

DOI: 10.3303/CET1335117

CO_2 Emission Reduction in the Cement Industry

Hrvoje Mikulčić[a,*], Milan Vujanović[a], Nataša Markovska[b], Risto V. Filkoski[c], Marko Ban[a], Neven Duić[a]

[a] Faculty of Mechanical Engineering and Naval Architecture, University of Zagreb, Ivana Lučića 5, 10000 Zagreb, Croatia
[b] Research Center for Energy, Informatics and Materials, Macedonian Academy of Sciences and Arts, Krste Misirkov 2, 1000 Skopje, Macedonia
[c] Faculty of Mechanical Engineering, University "Ss Cyril and Methodius", Karpos II nn, 1000 Skopje, Macedonia
hrvoje.mikulcic@fsb.hr

The cement industry is one of the largest carbon emitting industrial sectors in the European Union (EU) and in the world. In line with the EU commitment to combat climate change, the cement industry needs to decrease significantly carbon emission. The cement manufacturing process is not only a source of combustion related CO_2 emissions, but it is also a large source of industrial process related CO_2 emissions. There are several effective measures which can be applied in cement manufacturing processes to achieve emissions reduction targets. Simultaneously, these measures can reduce the local environmental impacts and improve the competitiveness of the cement industry. In this paper, the following measures for CO_2 emission reduction were analyzed in order to identify their environmental effectiveness: use of alternative fuels, more efficient kiln process, and co-production of synthetic fuels. The study was done on the case of a Macedonian cement plant. It was confirmed that the implementation of the selected mitigation measures results in substantial CO_2 emission reduction.

1. Introduction

During cement production process large amounts of different greenhouse gases, especially CO_2, are emitted. The cement industry alone accounts for approximately 4.1 % of EU's, and around 5 % of world's anthropogenic CO_2 emissions (Mikulčić et al., 2013). In line with the EU commitment to combat climate change, the cement industry, as the third largest carbon emitting industrial sector in EU, needs a more sustainable future. In 2005 the EU Emissions Trading Scheme (EU ETS) was launched. Due to high CO_2 emissions from the cement manufacturing process, cement plants within the EU are obliged to participate in this trading scheme. Cement manufacturing process is not only a source of combustion related CO_2 emissions, but it is also a large source of industrial process related CO_2 emissions. The calcination and the combustion of fossil fuels are the main processes contributing to almost 90% of CO_2 emitted from the cement manufacturing. The remaining 10 % comes from the transport of raw material and some other production activities. The calcination process in fact, is the thermal decomposition of limestone into lime, needed for the production of clinker. Combustion of fossil fuels contributes to around 40 % of CO_2 emissions.
There are four main cement production processes which have the highest influence on the final cement quality, fuel consumption, and pollutant formation. These processes are: raw material preheating, calcination process, clinker burning, and clinker cooling. Prior to the raw material preheating, the raw material is collected, crushed, mixed with additives and transported to the cyclone preheating system. The cyclone preheating systems have been developed to improve the heat exchange process. Preheating occurs prior to the cement calciner and the rotary kiln. Once preheated, the raw material enters the cement calciner, where the calcination process occurs. Clinker burning occurs after the calcination process. It is the most energy demanding process in cement production. The temperature of 1450 ˚C ensures the clinker formation. After the clinkering process in the rotary kiln is finished, the cement clinker is cooled

Please cite this article as: Mikulcic H., Vujanovic M., Markovska N., Filkoski R., Ban M., Duic N., 2013, Co2 emission reduction in the cement industry, Chemical Engineering Transactions, 35, 703-708 DOI:10.3303/CET1335117

down to 100-200 ˚C. The cooling is rapid, preventing thus undesirable chemical reactions (Mikulčić et al., 2012a).

There are several effective measures, which can be applied in cement manufacturing processes to achieve CO_2 emissions reduction. Simultaneously these measures can reduce the local environmental impacts and improve the competitiveness of the cement industry. As the largest CO_2 emitter, the calcination process is the best to start with. The only way to reduce CO_2 emissions from the calcination process is to use alternative raw materials, which do not contain carbonates in their mineral structure. However, till now no economically viable minerals from which the produced cement is comparable by quality to the current portland-based cements, have been found (Gartner, 2004). Rosković and Bjegović (2005) studied the influence of the substitution of a part of clinker with mineral additions on the mechanical characteristics of cement and the reduction of CO_2 emissions. The study showed that by reducing clinker to cement ratio with various additives, the consumption of raw materials, thermal and electric energy, and the CO_2 emissions can be reduced notably. Due to high CO_2 content in the flue gases, the most effective way to reduce CO_2 emissions from the cement manufacturing process is to capture CO_2 from the flue gases and store it (Deja et al., 2010). Barker et al. (2009), based on a newly built cement plant in Scotland, United Kingdom, analysed the technologies that could be used for CO_2 capture in cement plants, their costs and barriers to their use. The study concluded that the oxy-combustion in contrast to the post-combustion is an economically better solution for CO_2 capture in cement plants, but still research and development is needed, in order to enable this technology to be deployed. In addition to the previously mentioned Carbon capture and storage (CCS) technologies, Bosoaga et al. (2009) analysed the use of amine scrubbing and the calcium looping technology, as potential CCS technologies in cement industry. The study showed that the benefit of the calcium looping technology is that the lime removed from the cycle could be used for the production of clinker, and therefore reduce the CO_2 emissions from the cement industry. Furthermore, by improving the energy efficiency of the clinker production process CO_2 emissions can also be reduced. The most energy efficient cement production technology, best available technology, today is the use of a dry rotary kiln together with a multi stage cyclone preheater and a calciner (Mikulčić et al., 2013). In the study of Ke et al. (2012), on the case of China's cement industry, it was shown that the energy efficiency of the cement production process will be crucial for the reduction of CO_2 emissions and energy consumption. Following the fact that the use of the best available technology can reduce CO_2 emissions, Moya et al. (2011) analysed the energy technology improvements that can contribute to the decrease of energy consumption and CO_2 emissions in the EU's cement industry. In the study of Valderrama et al. (2012), a life-cycle assessment methodology was used to compare the newly installed best available technology and the former clinker production line. The study showed that an environmental improvement was achieved by using the best available technology for the clinker manufacturing, in a form of less fuel consumption. Furthermore, Mikulčić et al. (2012b), by using suitable numerical models (Mikulčić et al., 2012c), presented the potential of computational fluid dynamics (CFD) to support the design and optimization of cement calciners operating conditions, and to support the reduction of CO_2 emissions from the cement manufacturing process.

However, it was found that the substituting fossil fuels with alternative fuels may play a major role in the reduction of CO_2 emissions. Fodor and Klemeš (2012) presented the use of waste as an alternative fuel and discussed the advantages and disadvantages of the current and the developing waste-to-energy technologies. The study showed how the different waste-to-energy technologies are being developed and analyses their potential for greenhouse gas (GHG) emissions reduction. Kääntee et al. (2004) analysed the use of alternative fuels in the cement industry, providing useful data for the optimization of the pyroprocessing process when alternative fuels are used. Mokrzycki et al. (2003) reported the advantages, both economical and environmental, of using alternative fuels in Polish cement plants. The study showed that the use of alternative fuels is an environmentally friendly method of waste utilization. Aranda-Uson et al. (2012) studied the use of sewage sludge as an alternative fuel in cement industry. The study showed that significant technical and environmental improvements in the cement production process can be achieved when the sewage sludge is used as an alternative fuel. Furthermore, Aranda-Uson et al. (2013) presented the use of alternative fuels and raw materials in the cement industry. The study showed that alternative fuels can decrease cement industry's environmental impact, and furthermore that it can lower the consumption of energy and material resources, and reduce the economic costs of cement production. Aside from the studies investigating the environmental potential of using alternative fuels in the cement industry, Mislej et al. (2012) studied both the combustion behaviour and the environmental effect of using alternative fuels for heat generation in cement kilns. The study showed that there is a great potential, especially environmental, of using alternative fuels in cement production.

All of these studies showed that by optimizing existing cement production lines, using more efficient technologies, installing CCS technologies, using alternative fuels and by reducing the clinker to cement

ratio, a reduction in CO_2 emissions can be achieved. However, none of these studies show the potential of combining renewable energy resources and cement manufacturing process. The co-production of synthetic fuel, a CO_2 emission reduction measure considered in this study, is a method that could show that potential. In this way, CO_2 emissions related to fuel combustion could be avoided.
In this study cement plant Usje was selected as the case study. The paper analyses the existing pyro-processing unit and its CO_2 emissions reduction potential. Three measures, which can be applied in the cement manufacturing processes, were analyzed in order to identify their environmental effectiveness. The implementation of these measures results in a significant decrease of CO_2 emissions in 2020, contributing to a more sustainable cement production.

2. Methodology

Direct CO_2 emissions from the production of cement are attributed to: (1) Calcination process - the process of transforming raw materials into clinker which is the main component of cement; (2) Fuel combustion - fuels (oil, coal, petrol coal etc.) burn in the kilns and produce CO_2 as a result of the chemical reaction between carbon and oxygen. Indirect emissions of CO_2 are released during the generation of electricity required for the production of clinker and cement, as well as during the transportation of raw materials, fuel and final products.
There are several measures in the cement industry, which can reduce CO_2 emissions significantly. One of the measures is the reduction of clinker to cement ratio with different additives. However, since this ratio for the final products of the cement plant Usje is already low, (amounts 0.57), it is not likely that its further reduction will be considered if blended cements, with at least as good performance and durability characteristics as the current portland-based cements, are to be produced. Furthermore, replacing fossil fuels with alternative fuels may play a major role in the reduction of CO_2 emissions. The other advantage of the application of this measure is the reduction of energy costs of cement production, and even more significant is the advantage that this measure is also an environmentally friendly method of waste utilization. Improving the energy efficiency of the kiln process is also one of the possibilities for CO_2 emissions reduction. Finally, co-production of synthetic fuel is a measure that could combine renewable energy resources and cement manufacturing process, and recycling the CO_2 from the flue gases lower CO_2 emissions notably.
Most of these measures are influenced to a large extent by environmental policy and legal framework and integration of these measures will only be possible if the policy framework will foster the cost-effective deployment of the best available technology. In this study, the CO_2 emissions for each of the named measure were calculated according to the Intergovernmental Panel on Climate Change (IPCC) methodology (IPCC, 2000). This methodology is widely used for tracking and reporting of greenhouse gas emissions from the industrial facilities. For cement industry it specifies that there are two different sources of CO_2 emissions: the combustion of fossil fuels, and the thermal decomposition of limestone, known as the calcination process. The former refers to combustion CO_2 emissions, and the latter refers to process CO_2 emissions.

3. Results and discussion

The case study of this paper is the cement plant Usje, Macedonia's only cement plant. The Republic of Macedonia, a party of the United Nations Framework Convention on Climate Change (UNFCCC) since 1998, belongs to the non-Annex I countries (group of countries without binding targets), but as a candidate country for EU membership, it is committed to develop inventory of GHG, climate change mitigation and adaptation plans, to undertake climate change related observation and monitoring, research and development, education and public awareness, as well as to report regularly the national climate change activities in National Communications (Taseska et al., 2008).
The kiln process applied in the cement plant Usje is a dry rotary kiln, with four-stage cyclone pre-heater and a clinker cooler. Calciner is not used in this cement plant; however the plant operator is planning to increase the cement production while decreasing the CO_2 emissions. To ensure both criteria are satisfied, the plant operator plans to improve the kiln process by including the calciner in it.
The energy efficiency of a particular cement plant is evaluated in a way that the specific energy consumption of that particular cement plant is compared with the specific energy consumption of a benchmark. The specific energy consumption is also used to evaluate the improvements in the energy efficiency of the cement production process (Ali et al., 2011). The current average specific thermal energy consumption of a kiln process in the Usje cement plant is 3.7 GJ/t clinker. It can be noted that there is still space for certain improvement of the energy efficiency of the Usje cement plant, and that the next step in

improving the energy efficiency would be the addition of a calciner prior to the rotary kiln. Some energy consumption indicators of the Usje cement plant are shown in Table 1.

Table 1: Energy consumption indicators

Electricity consumption for production of clinker	80 GWh/y
Electricity consumption for production of cement	73.5 GWh/y
Specific consumption of electricity for clinker	80 kWh/t clinker
Specific consumption of electricity for cement	42 kWh/t cement
Fuel consumption per kg clinker	3,700 kJ/kg clinker
Total fuel consumption	3,700x109 kJ/y
Electricity consumption for other activities in the plant	40 GWh/y

The CO_2 emissions reduction measures considered here are: (a) use of alternative fuels; (b) more efficient kiln process; (c) co-production of synthetic fuels. The selection of the measures was based on the criteria of the ability to decrease CO_2 emissions compared to current practice, prospects for realisation, and level of difficulty of implementation.

To compare the environmental effectiveness of each measure, the CO_2 emissions for last five years from the cement plant Usje were calculated. The results (see Table 2) show that CO_2 emissions from cement manufacturing in Macedonia, decreased sharply from 2007 until 2009, due to the decreased cement production during the economic crisis, and after 2009 when Macedonia's economy started to recover, the CO_2 emissions from the cement production slowly increase.

Table 2: CO_2 emissions from the Usje cement plant

Year	Emissions of CO_2, ktCO_2
2007	867
2008	814
2009	439
2010	442
2011	457

3.1 Use of alternative fuels

The alternative fuel used for the calculation of the potential CO_2 emissions reduction was the refused derived fuel well known as RDF. RDF is produced from the high calorific part of the waste after the removal of the inert substances and drying of the waste. By using RDF together with fossil fuels, a decrease in CO_2 emissions can be achieved, due to biogenic part of the RDF. For the purpose of this study, it was assumed that the biogenic part (wood, textile, paper, etc.) in RDF is 40 % (Schneider et al., 2012).

The CO_2 emissions reduction potential was calculated by estimating the total energy consumption of the cement factory. An assumption was made that RDF has the lower heating value of 15 MJ/kg and that in 2020 RDF will have a 50 % share in the fuel mix that will be used for the cement production. The actual amount of the RDF, that will be used in 2020 as the alternative fuel is 123.5 kt. That results in the CO_2 emission reduction potential of 104.4 ktCO_2eq in 2020.

3.2 More efficient kiln process

For more efficient kiln process it was assumed that the existing kiln process, with an average specific thermal energy consumption of 3.7 GJ/t clinker, will be replaced with the currently best available kiln process that has an average specific thermal energy consumption of 3.0 GJ/t clinker. The direct result of the implementation of a more efficient kiln process is the decrease in fuel consumption. The actual amount of the fossil fuel that will be used in 2020 is 89.5 kt for a more efficient kiln process, while 110.4 kt of fossil fuel will be needed for the existing kiln process. The CO_2 emissions reduction potential in 2020 for a more efficient kiln process is 65.7 ktCO_2eq.

3.3 Co-production of synthetic fuels

The use of synthetic hydrocarbon fuels in cement production processes is a possible solution for reducing fuel consumption and CO_2 emissions. The basis for producing synthetic hydrocarbon fuels is the synthetic gas, or shortly syngas, a gas mixture that contains varying amounts of CO and H_2. The syngas is produced during the co-electrolysis of CO_2 and H_2O in solid oxide electrolysis cells. After that, the Fischer–Tropsch synthesis can be used to produce liquid hydrocarbon fuel from the syngas. In this study an assumption was made that the sources of CO_2, the basis for hydrocarbon fuels, is the post-combustion carbon capture and recycling (CCR) technology. The electricity which enables the co-electrolysis process is provided by wind turbines. This option is chosen to integrate renewable resources into the cement production process. Furthermore it was assumed that due to modest efficiency of current solid oxide electrolysis cells and the Fischer–Tropsch synthesis, methane, the produced synthetic fuel, will have a 20 % share in the fuel mix in 2020.

The CO_2 emission reduction potential was calculated by estimating the total energy consumption of the cement factory. For methane the lower heating value of 50 MJ/kg was used. The actual amount of the methane, that will be used in 2020 as the synthetic fuel is 14.8 kt. That results in the CO_2 emission reduction potential of 28.8 ktCO_2eq in 2020.

Table 3 summarizes the CO_2 reduction potential for three considered measures. It can be concluded that the use of alternative fuels has the biggest CO_2 reduction potential. Hence, this measure should be first for the implementation in cement manufacturing processes in the cement plant Usje.

Table 3: CO_2 emissions reduction potential

CO_2 emissions reduction measure	CO_2 emissions reduction potential, ktCO_2eq
Use of alternative fuels	104.4
More efficient kiln process	65.7
Co-production of synthetic fuels	28.8

4. Conclusion

Reduction of CO_2 emission in cement industry is one of the most important measures for achieving the EU climate targets for 2020 and beyond. The paper analyses the environmental effectiveness of three different CO_2 emissions reduction measures, in order to determine the priorities for implementation in the considered case. The three considered measures were: Use of alternative fuels, More efficient kiln process, and Co-production of synthetic fuels. It should be noted that the recycling of CO_2 into synthetic fuels will open the door to renewable energy in the cement industry sector.

The integral CO_2 emissions reduction potential of the three measures shows that approximately 0.2 million tons of CO_2 can be avoided in 2020, which is around 1.7 % of Macedonia's current GHG emissions, or around 40 % of total CO_2 emissions of the cement plant Usje. These figures also show that the CO_2 emission reduction potential in cement industry could be a significant part of the efforts to meet the potential post-Kyoto target, and that the implementation of the considered mitigation measures contributes to more sustainable cement production.

References

Ali M.B., Saidur R., Hossain M.S., 2011, A review on emission analysis in cement industries, Renew. Sustain. Energy Rev. 15, 2252-2261.

Aranda-Usón A., Ferreira G., López-Sabirón A.M., Sastresa E.L., de Guinoa A.S., 2012, Characterisation and environmental analysis of sewage sludge as secondary fuel for cement manufacturing, Chem. Eng. Trans. 29, 457-462.

Aranda-Usón A., López-Sabirón A.M., Ferreira G., Sastresa E.L., 2013, Uses of alternative fuels and raw materials in the cement industry as sustainable waste management options, Renew. Sustain. Energy Rev. 23, 242-260.

Barker D.J., Turner S.A., Napier-Moore P.A., Clark M., 2009, Davison JE. CO_2 capture in the cement industry. Energy Procedia 1, 87-94.

Bosoaga A., Masek O., Oakey J.E., 2009, CO_2 capture technologies for cement industry, Energy Procedia 1, 133-140.

Deja J., Uliasz-Bochenczyk A., Mokrzycki E., 2010, CO_2 emissions from Polish cement industry. Intern. J. Greenh. Gas Control 4, 583-588.

Fodor Z., Klemeš J.J., 2012, Waste as alternative fuel – Minimising emissions and effluents by advanced design, Process Saf. Environ. Prot. 90, 263-284.

Gartner E., 2004, Industrially interesting approaches to "low-CO_2" cements, Cem. Concr. Res. 34, 1489-1498.

IPCC (Intergovernmental Panel on Climate Change), 2000, Good Practice Guidance and Uncertainty Management in National Greenhouse Gas Inventories <www.ipcc-nggip.iges.or.jp/public/gp/english/> accessed: 21.03.2013

Kääntee U., Zevenhoven R., Backman R., Hupa M., 2004, Cement manufacturing using alternative fuels and the advantages of process modelling, Fuel Process. Technol. 85, 293-301.

Ke J., Zheng N., Fridley D., Price L., Zhou N., 2012, Potential energy savings and CO_2 emissions reduction of China's cement industry, Energy Procedia 45, 739-751.

Mikulčić H., Vujanović M., Fidaros D.K., Priesching P., Minić I., Tatschl R., Duić N., Stefanović G., 2012a, The application of CFD modelling to support the reduction of CO_2 emissions in cement industry, Energy 45, 464-473.

Mikulčić H., von Berg E., Vujanović M., Priesching P., Tatschl R., Duić N., 2012b, CFD analysis of a cement calciner for a cleaner cement production, Chem. Eng. Trans. 29, 1513-1518.

Mikulčić H., von Berg E., Vujanović M., Priesching P., Perković L., Tatschl R., Duić N., 2012c, Numerical modelling of calcination reaction mechanism for cement production, Chem. Eng. Sci. 69, 607-615.

Mikulčić H., Vujanović M., Duić N., 2013, Reducing the CO_2 emissions in Croatian cement industry, Appl. Energy 101, 41-48.

Mislej V., Novosel B., Vuk T., Grilc V., Mlakar E., 2012, Combustion behaviour and products of dried sewage sludge – prediction by thermogravimetric analysis and monitoring the co-incineration process in a cement factory, Chem. Eng. Trans. 29, 685-690.

Mokrzycki E., Uliasz-Bocheńczyk A., Sarna M., 2003, Use of alternative fuels in the Polish cement industry, Appl. Energy 74, 101-111.

Moya J.A., Pardo N., Mercier A., 2011, The potential for improvements in energy efficiency and CO_2 emissions in the EU27 cement industry and the relationship with the capital budgeting decision criteria, J. Clean. Prod. 19, 1207-1215.

Rosković R., Bjegović D., 2005, Role of mineral additions in reducing CO_2 emission, Cem. Concr. Res. 35, 974-78.

Scheider D.R., Kirac M., Hublin A., 2012, Cost-effectiveness of GHG emission reduction measures and energy recovery from municipal waste in Croatia, Energy 48, 203-211.

Taseska V., Markovska N., Causevski A., Bosevski T., Pop-Jordanov J., 2011, Greenhouse gases (GHG) emissions reduction in a power system predominantly based on lignite, Energy 36, 2266-2270.

Valderrama C., Granados R., Cortina J.L., Gasol C.M., Guillem M., Josa A., 2012, Implementation of best available techniques in cement manufacturing: a life-cycle assessment study, J. Clean. Prod. 25, 60-67.