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Reducing greenhouse gasses emissions by fostering the deployment of alternative raw materials and energy sources in the cleaner cement manufacturing process

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ABSTRACT

The cement production industry worldwide is one of the largest CO_2 emitting industrial sectors. It accounts for a considerable amount of total global greenhouse gas (GHG) emissions. Due to the increasing awareness of global warming, more energy efficient cement production is increasingly being emphasized. One of the priorities is to reduce the energy demand and innovate the production process to move towards the cleaner production as: Energy efficiency improvements; Waste heat recovery; Reduction of clinker/cement ratio and use of alternative raw materials; Substitution of fossil fuels with alternative energy sources. When the GHG emissions at source opportunities are close to being exhausted, the other mitigations options should be considered such as: CO_2 capture and storage. This is however in most cases not the final solution from the point of Life cycle assessment (LCA). In recent years various mitigation measures are gaining on the importance and the cement industry is more and more shifting to cleaner production. Among the others, there are two measures, which can reduce the GHG emissions considerably: the use of alternative raw materials and alternative fuels. The challenge for the cement industry is to use alternative raw materials especially those originating from other industries where they are considered as by-products or even waste. Some of these by-products include: Bottom ash from municipal solid waste incinerators; Fly ash from coal power plants; Gypsum from the desulfurization plants used in power plants. Another important measure is the energy efficiency improvement in existing cement plants. There are various approaches for controlling and improving the energy efficiency within existing cement manufacturing units, however, mathematical modelling, simulation, optimisation and Process Integration are increasingly gaining in importance. The mathematical modelling approach uses the numerical simulations for the investigation of the thermo-chemical processes occurring inside of the manufacturing unit. The results gained are being used to enhance the efficiency of cement production. They improve the understanding of the flow characteristics and transport phenomena taking place inside the cement combustion unit. The objective of this paper is to review the current status of the cleaner cement manufacturing, the cement industry's shifting to alternative raw materials and alternative energy sources, and the modelling of the thermo-chemical processes inside the cement combustion units. Additionally, some critical issues, which up to now have not been adequately resolved, are outlined.

1. Introduction

The recent Fifth Assessment Report of the Intergovernmental Panel on Climate Change - IPCC (IPCC, 2014) states that recent anthropogenic emissions of greenhouse gases - GHG are higher than ever and that the influence of mankind on the global climate changes is clear. Despite a growing number of climate change

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mitigation policies, the anthropogenic GHG emissions in 2013 were highest in history and reached 49 ± 4.5 $GtCO_2$-eq. Out of this figure, CO_2 emissions coming from fossil fuel combustion and industrial processes and from forestry and other land use represented 76%. The actual CO_2 concentrations in the atmosphere have increased significantly from of the pre-industrial era to the year 2013. The International Energy Agency – IEA (2014) reports that the CO_2 concentration was increased from pre-industrial value of 280 ppm, to the year 2013 record high CO_2 concentration of 396 ppm. The main contributors to the increase of CO_2 emissions and its concentration in the atmosphere are population growth, standard of living and economic growth. Out of 214 countries today, two countries and one trans-national organization are responsible for 55% of total global CO_2 emissions (Olivier et al., 2014). China generates 10.3 Gt CO_2 emissions or 29%, the United States are responsible for 5.3 Gt CO_2 or 15%, and the European Union - EU28 is responsible for 3.7 Gt CO_2 or 11% of total global CO_2 emissions. However the responsibility should consider virtual footprint flows related to the export and import, which make the figures slightly different, some overview has been offered by (Yong et al., 2016).

Development of footprint assessment techniques over the last decade has provided a set of tools for monitoring CO_2 emissions and water flows in the world (Čuček et al., 2012a). An overview of the virtual CO_2 and virtual water flow trends in the international trade based on consumption perspective was performed by Liu et al. (2015) and the results of this study are shown in Fig. 1. This paper on the base of the recent literature indicates that:

(1) There are significant CO_2 gaps between producer's and consumer's emissions, and USA and EU have high absolute net imports CO_2 budget.
(2) China is an exporting country and increasingly carries a load of CO_2 emission and virtual water export that are triggered due to consumption in other importing countries.
(3) By imported products that are produced with lower carbon emission intensity and less water consumption then in the domestic industry, international trade can reduce global environmental pressure.

Liu et al. (2015) concluded that a future direction should be focused into two main areas:

(1) To provide the self-sufficient regions based on more efficient processes by combining production of surrounding countries.
(2) To develop the shared mechanism and market share of virtual carbon and virtual water between trading partners regionally and internationally.

However, they are some very significant sources of GHG out of the industry. In 2015, more than 94,000 fires, most in carbon-rich peat lands, have engulfed the island nation of Indonesia, sending thick, acrid smoke into the air (Patterson, 2015). The haze is affecting the health of millions of people there in Malaysia and Singapore. As a result, immense stores of carbon have been released by the fires into the atmosphere. By late October 2015 the fires have released an estimated 10^9 t of CO_2, about 3% of global fossil fuel emissions or more carbon emissions than released by Germany in 2013, according to calculations by van der Werf (2015). Or another way to put it, since September 2015, on 26 occasions, daily emissions from Indonesia's fires exceeded daily emissions from the entire US economy, which is 20 times larger than Indonesia.

When it comes to the sectoral analysis of the origin of CO_2 emissions, it is known that electricity and heat generation, transport, and industry sector emit over 80% of global CO_2 emissions (Benhelal et al., 2013). Due to this reason, there is a great need for the reduction of CO_2 emissions coming from these sectors (Klemeš et al., 2010) and with more recent date (Klemeš et al., 2012). In

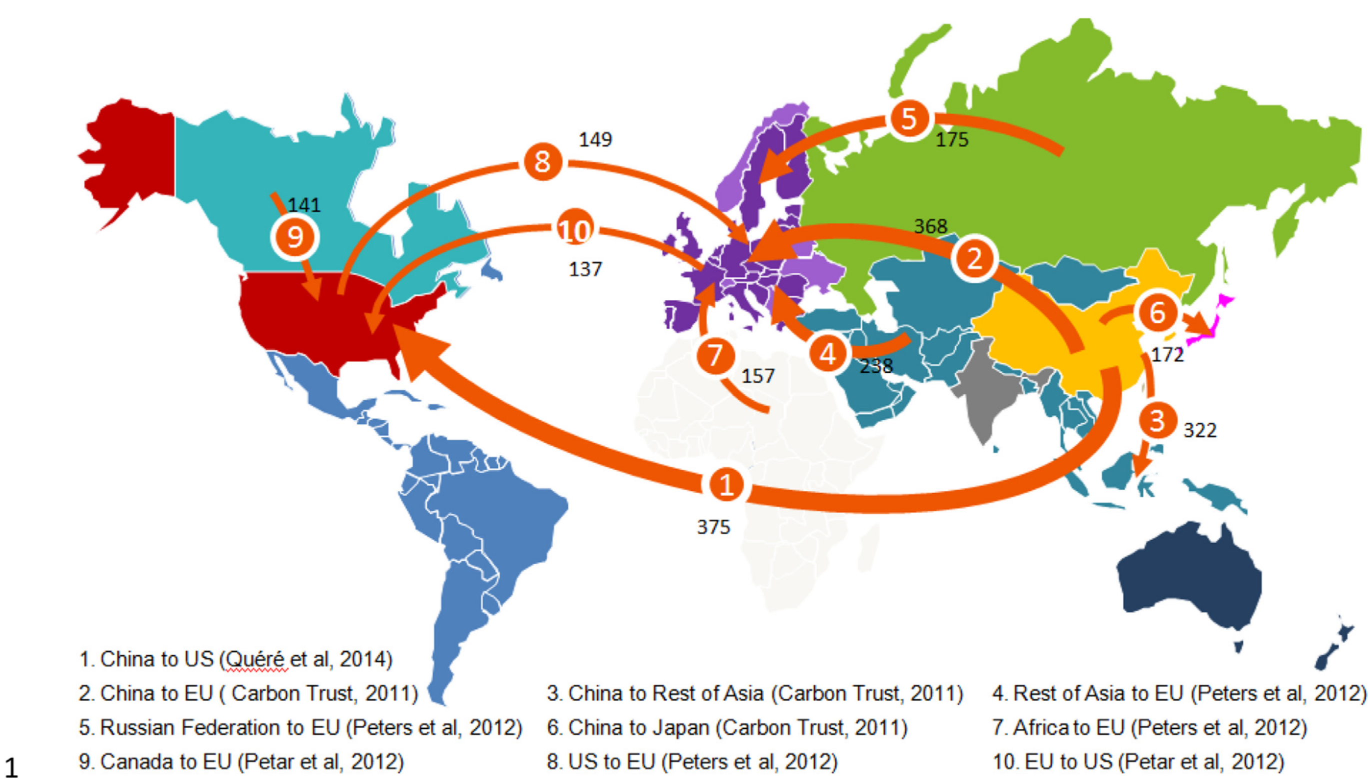

Fig. 1. Ten largest inter-regional flows of embodied CO_2 emissions in 2014 (Mt CO_2) (after Liu et al., 2015).

response to this situation many research initiatives for increasing of the efficiencies of various production processes have already been conducted (Dovì et al., 2009).

Cement industry as an energy intensive industry is alone responsible for a large amount of CO_2 emissions. Global cement production grew by over 73% between 2005 and 2013 from 2310 Mt to 4000 Mt, meaning that there was also a considerable increase in CO_2 emissions from cement production (CEMBERAU, 2014). Thus, the cement industry is an important industrial polluter in terms of GHG, and emission reductions in this sector may lead to significant decreases of overall GHG releases (Valderrama et al., 2012).

In the reported literature, there are differences in the estimation of the CO_2 emissions related to cement production. This difference in the estimation can be observed in Table 1. When all of the studies are summarized cement production accounts for roughly 5–8% of global CO_2 emissions.

However, to partly set this figure right, if one assumes cement production generates a world-averaged carbon emission of 0.83 kg CO_2/kg cement produced (Teklay et al., 2015), multiplies it with the produced cement (Oh et al., 2014), and compares it to the total CO_2 emissions (IPCC, 2014), gets that cement production contributes up to 8% of total global anthropogenic CO_2 emissions. This estimation is in correlation with the latest report on global CO_2 emission trends by Olivier et al. (2015). Meaning that cement industry alone, has a significant and growing, with the escalation demand for the concrete constructions, impact on the environment.

The growing anthropogenic GHG emissions and increasing global demand for cement are general drivers that motivate finding solutions for managing GHG emissions in the cement industry. The objective of this paper is to review the current status and latest literature on the cement production, the cement industry's shifting to alternative raw materials and alternative energy sources and the modelling of the thermo-chemical processes inside the cement combustion units. Additionally some critical problems, which up to now have not been adequately resolved, are outlined.

2. Method of the systematic literature review and the literature analysis

This paper offers a systematic literature review and a bibliometric analysis of cement industry papers published in the past 5 years. Due to the extensive and timely coverage of over 20,000+ journals from the main publishers of peer-reviewed papers, like ACS Publications, Elsevier, Emerald, Informs, Inderscience, Springer, Taylor and Francis and Wiley, the Scopus database has been chosen for literature search. The search has been focused to peer-reviewed papers and reviews published in journals in order to collect high quality research papers. The database search used the following keywords, in pseudo-code: 'TITLE-ABS-KEY (cement industry) AND DOCTYPE (ar OR re)'. In the following the results presented were analysed with Scopus online analysis tools.

Table 1
Different estimations of the CO_2 emissions related to cement production, compared to global annual anthropogenic CO_2 emissions.

5% of anthropogenic CO_2 emissions	7% of anthropogenic CO_2 emissions	5–8% of anthropogenic CO_2 emissions
Marques and Neves-Silva, 2014;	Ali et al., 2011;	Habert et al., 2011;
Mikulčić et al., 2013b;	Deja et al., 2010;	Huntzinger and Eatmon, 2009;
Pelisser et al., 2012;	Li et al., 2013;	Kajaste and Hurme, 2015;
Sjølie, 2012	Oh et al., 2014	Van Deventer et al., 2012

The search for relevant cement industry papers shows that in period from 1945 till 2016 there have been 6873 papers published. However, the majority of papers about cement industry have been published in last 20 years. From 1995 till 2016, 5816 papers dealing with cement industry have been published. This can be clearly observed when looking at Fig. 2, where the evolution of the number of cement papers over the years is shown. Fig. 2 shows that some 20 years ago, a sharp increase in published papers occurred, meaning that at that time the importance of cement industry was recognized and that the research related to the cement industry increased. What can also be observed is the decrease of number of papers related to cement industry during the economic crisis from 2007 till 2010, which had a major effect on the cement industry sector as well.

Regarding the sources of the papers, there are a large number of journals that cover the topic of cement industry. However, for some of these journals coverage discontinued in Scopus, and therefore these journals are not shown in Fig. 3. In Fig. 3 five journals that published the largest number of papers related to cement industry are shown. As can be seen Journal of Cleaner Production is the only journal among the first five journals that has a steady increase in the number of published papers during last 10 y. Other journals show fluctuations and decrease in the number of published papers related to cement industry over the last ten years. One of the strong reasons for the increasing position of Journal of Cleaner Production is the increasing demand for cleaner cement production with the reduction of the environmental impact represented by footprints. When it comes to most prolific authors in this field in last three years, the list of the five most prolific authors in this field is presented in Fig. 4.

In the following sections the most relevant and cited papers are analysed regarding their connection to the topic of this review paper.

3. Cement manufacturing process

3.1. Brief process description

The best available technology, the one with the lowest energy consumption, for the cement manufacturing today, is the use of a rotary kiln together with multi-stage cyclone preheater system and a calciner. Fig. 5 illustrates the stages of the cement production. There are four sub-processes that have the most influence on final cement quality and fuel consumption, namely: raw material preheating, calcination, clinker burning, and clinker cooling (Fidaros et al., 2007). 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 enhance the heat exchange between the raw material and the flue gases. The preheating takes place prior to the calciner and the rotary kiln and can have several stages, depending on how many cyclones are used. At each stage of the preheating system, e.g. in each cyclone, the principle of the heat exchange is the same. Raw material is heated by moving counter to the flow of the hot flue gases coming from the rotary kiln. This counter-flow movement effect is due to the particle separation phenomena occurring within the gas cyclones. The separation of the solid particles from the gas is done by the highly tangential flow entering the cyclone. The centrifugal force acting on the particles directs them to the wall, separating them from the flow, and due to the gravitational force the particles slide to the lower part of the cyclone. In contrast to the solid particles the gas flow has a different behaviour. Firstly the gas swirls downwards in the outer cyclone part, where the separation is done, and then in the lower part of the cyclone where the axial

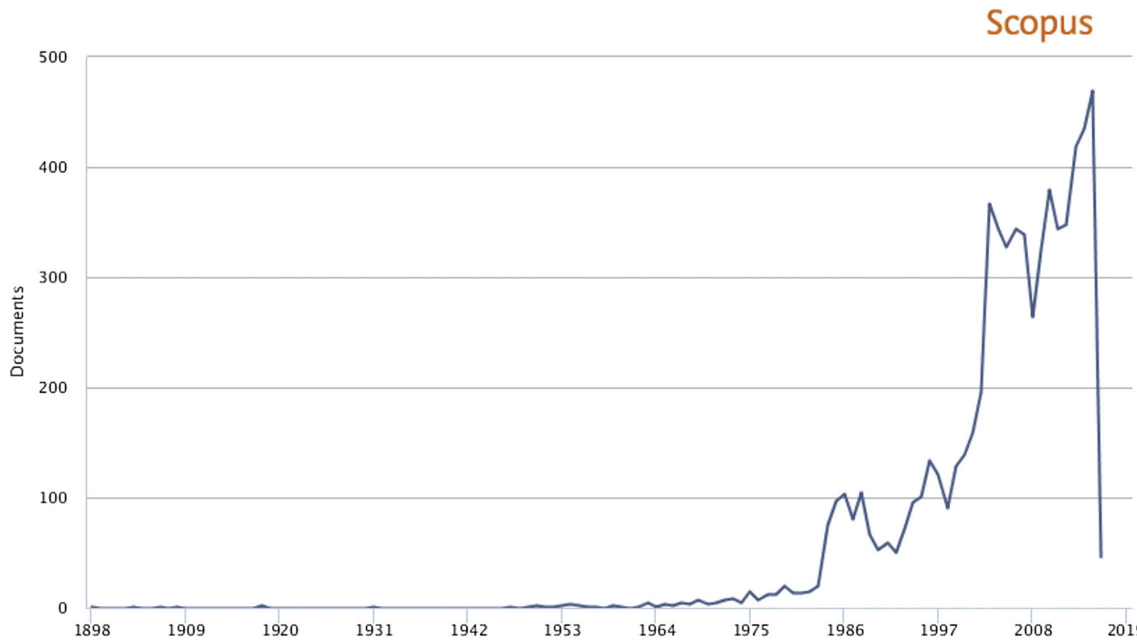

Fig. 2. Cement papers over the years (extracted from SCOPUS, 2016).

velocity reverses, the gas starts to swirl upwards in the inner cyclone region. This process is repeated until the raw material goes through all the cyclones (Mikulčić et al., 2014b).

After preheating, raw material enters the cement calciner. Cement calciner, is a combustion unit found prior to the rotary kiln, and inside of it, the raw material, mainly composed of limestone, undergoes the calcination process. The calcination process is a strongly endothermic reaction that requires combustion heat released by the fuel, indicating that endothermic limestone calcination and exothermic fuel combustion proceed simultaneously (Chen et al., 2012). According to Szabó et al. (2006) a decrease of energy consumption by 8–11% can be achieved when a rotary kiln is used together with a calciner. This decrease is due to the fact that cement calciners have lower operating temperatures than rotary kilns. To ensure a temperature of 850 °C, needed for a stable calcination process, cement calciners use heat from the combustion of solid fuels along with the exhaust gases from a rotary kiln (Mikulčić et al., 2012b).

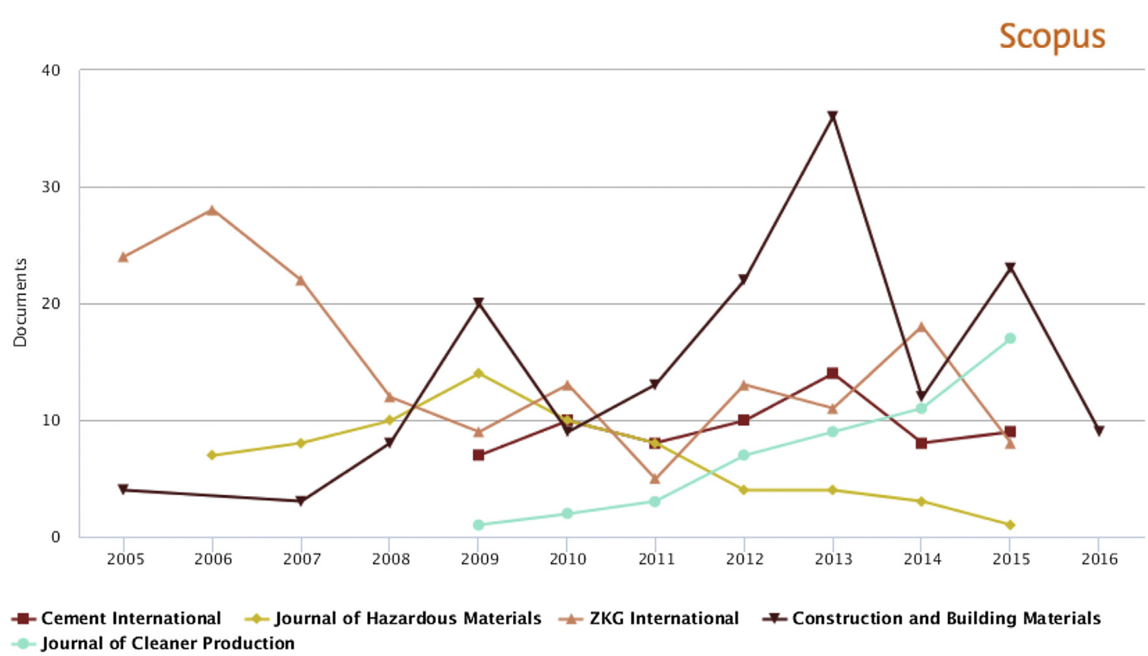

Fig. 3. Source of papers over the years (extracted from SCOPUS, 2016).

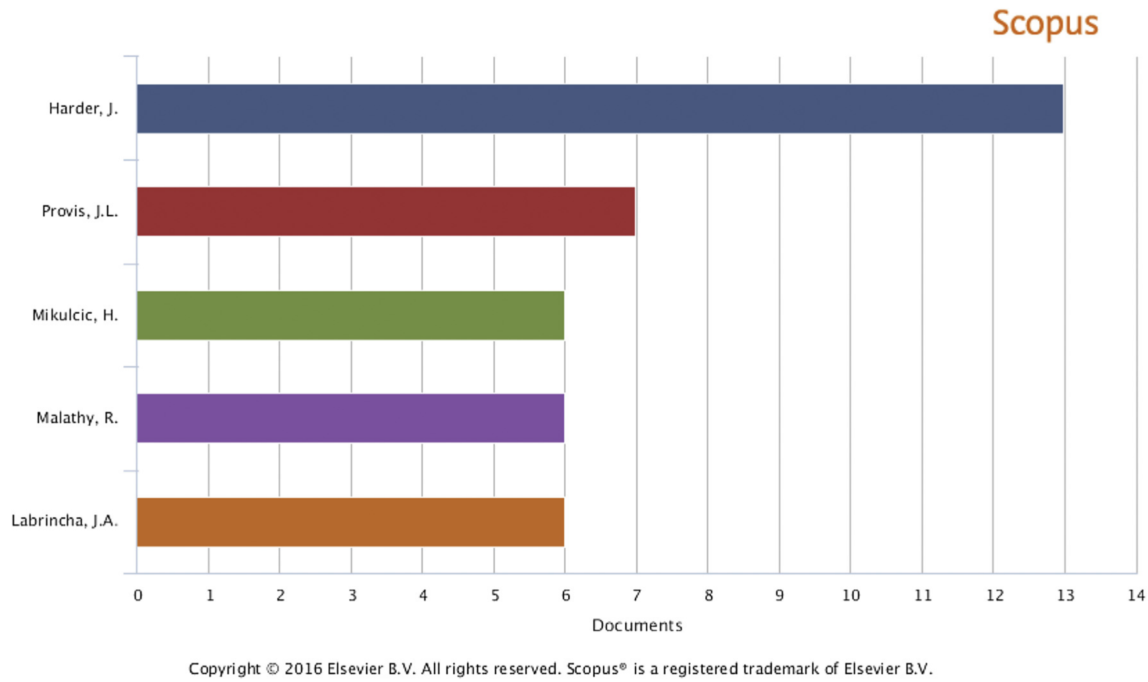

Fig. 4. Most publishing authors (extracted from SCOPUS, 2016).

Clinker burning is the highest energy demanding process in cement production. It occurs after the calcination process. The clinker is produced in a rotary kiln which rotates 3–5 times per minute, and is positioned at an angle of 3–4°. This angle causes the material to slide and tumble down through the hotter zones towards the flame. The temperature of 1450 °C ensures the clinker formation. After the clinkering process in the rotary kiln is finished, the cement clinker is rapidly cooled down to 100–200 °C (Ecofys, 2009). This process is done rapidly to prevent undesirable chemical reactions. Blending of clinker with different additives follows the clinker cooling process. At that point the composition of the final product - cement is obtained. Afterwards the cement is milled, stored in the cement silo, and distributed to consumers.

3.2. Worldwide production

Due to the significant environmental impact of cement production, over the past decades several mitigation measures have appeared, aiming mainly at the environmental conservation in terms of reducing CO_2 emissions. In recent years, there have been numerous studies worldwide discussing energy conservation policies, estimating the CO_2 mitigation potential, and considering

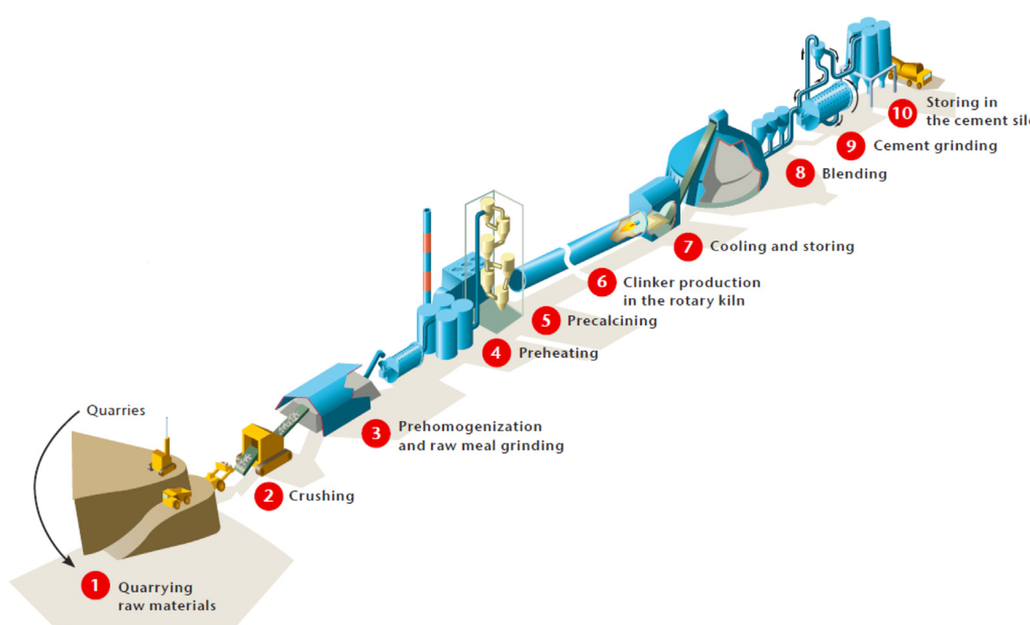

Fig. 5. Cement manufacturing process (IEA Cement Technology Roadmap, 2009).

Table 2
Peer-reviewed papers on the environmental impact of cement production at national and regional levels (SCOPUS, 2016).

Industrialized countries/territories	Peer-reviewed paper
EU	Moya et al., 2011; Pardo et al., 2011; Supino et al., 2016
Nordic cement industry	Rootzén and Johnsson, 2015
Spain	Castañón et al., 2015; García-Gusano et al., 2015
United States	Xu et al., 2013
Germany	Brunke and Blesl, 2014
Poland	Deja et al., 2010
South Korea	Suk et al., 2014
Japan	Oh et al., 2014
Developing countries	
China	Fujii et al., 2013; Hasanbeigi et al., 2013; Lei et al., 2011; Li et al., 2014; Tan et al., 2015; Wang et al., 2013; Wang et al., 2014; Wang et al., 2015; Xu et al., 2014; Zhang et al., 2015
Vietnam	Nguyen and Hens, 2013
South Africa	Swanepoel et al., 2014
India	Thirugnanasambandam et al., 2011; Morrow et al., 2014
Turkey	Ekincioglu et al., 2013
Iran	Ansari and Seifi, 2013; Ostad-Ahmad-Ghorabi and Attari, 2013
Bangladesh	Hoque and Clarke, 2013
Thailand	Hasanbeigi et al., 2010

technology evaluation for the cement industry. Some of these studies investigated the effect of mitigation measures at the global level, such as the study conducted by the International Energy Agency – IEA (IEA Cement Technology Roadmap, 2009). However, the majority of these studies evaluated the environmental impact of cement production at national and regional levels.

As can be seen from Table 2, due to the rapid economic growth and vast urbanization, the majority of the studies related to the cement industry are done for the developing countries in Asia, and

Table 3
Global cement production in 2012 (Mikulčić et al., 2016).

Country	Production (Mt)	Share in the world production
China	2150	58.1%
India	250	6.7%
United States	74	2.0%
Brazil	70	1.9%
Iran	65	1.8%
Vietnam	65	1.8%
Turkey	60	1.6%
Russian Federation	60	1.6%
Japan	52	1.4%
South Korea	49	1.3%
Egypt	44	1.2%
Saudi Arabia	43	1.2%
Mexico	36	1.0%
Germany	34	0.9%
Thailand	33	0.9%
Pakistan	32	0.9%
Italy	32	0.9%
Indonesia	31	0.8%
Spain	20	0.5%
Other (rounded)	500	13.5%
World total (rounded)	3700	–

especially for China. The reason for this is most easily seen in Table 3 where the global cement production for 2012 is given. Table 3 shows that the vast majority of cement production is located in developing countries, especially in Asia.

The importance of cement production in these developing economies can also be observed when comparing the annual CO_2 emissions from cement production in industrialised countries and developing countries. In the EU, the cement industry contributes to about 4.1% of total CO_2 emissions (Pardo et al., 2011). This share varies from one EU country to another, in Spain cement industry is responsible for 7% of Spanish CO_2 emissions (García-Gusano et al., 2015). In EU highly developed country Germany, this share is lower, and the cement industry accounts for 2.9% of Germany CO_2 emissions (Brunke and Blesl, 2014). This is similar for the cement industry in USA, where cement production is responsible for about 2% of total CO_2 emissions (Worrell and Galitsky, 2008). Whereas in the China, world's largest cement producing country and world's largest emitter of GHG emissions, 15% of total CO_2 emissions are related to cement production (Chen et al., 2014). All of the previously named studies stated that still there is a great challenge in attempting to approach sustainability in the cement industry.

3.3. Sources of CO_2 emissions from cement manufacturing

The cement and lime industries are unique due to the fact that the majority of greenhouse gas emissions are not caused by energy use from fuel combustion, but come from the raw materials themselves. The calcination process and the combustion of fossil fuels are the main processes contributing to high CO_2 emissions, where the first one contributes to around 50%, and the latter one contributes to almost 40% of CO_2 emitted from the cement manufacturing process. The remaining 10% comes from the transport of raw material and other manufacturing activities (Benhelal et al., 2012).

4. Pre-combustion and combustion CO_2 mitigation options

4.1. Reduction of clinker/cement ration and use of alternative raw materials

Waste-derived or by-product materials can be utilised to replace primary raw materials used in the cement clinker recipe. In order to contribute to lowering of energy consumption in clinker burning and reducing the associated CO_2 emission, suitable alternative materials should contain CaO; the presence of other major constituent oxides including SiO_2, Al_2O_3 and Fe_2O_3 is desirable. The use of alternative materials in clinker recipe requires a cautious approach as any alteration in cement's chemistry will affect the quality of the end product. The composition and properties of the clinker, as well as the end product must conform to technical specifications such as EN 197-1:2000 or EN 197-4:2004.

The reduction of clinker to cement ratio with different additives is another mitigation measure which can reduce CO_2 emissions significantly. However, since this measure has a direct effect on the performance and durability characteristics of the final products concrete special care needs to be taken that these blended cements, have at least as good performance and durability characteristics as the current Portland-based cements (Mikulčić et al., 2013b).

4.2. Substitution of fossil fuels with alternative energy sources

During the last decades, an interest in replacing fossil fuels with selected waste, biomass, and by-products with recoverable heating value, defined as alternative fuels, has arisen, in order to minimize production cost, reduce environmental impact, and reduce fuel costs. The utilization of alternative fuels imposes challenges on the calciner and kiln operation, because these fuels have different combustion characteristics, compared to fossil fuels. A particular concern is incomplete combustion of these fuels, since this may alter the behaviour of minor elements such as S, Cl, Na, and K. These elements are known to be circulating or volatile elements in the kiln system. Compounds containing these elements evaporate when exposed to high temperatures, and may subsequently condense in cooler parts of the plant (Cortada Mut et al., 2015).

4.3. Energy efficiency improvements

Specific energy consumption is a key indicator of the efficiency of a cement plant in its production of clinker (in GJ/t clinker). A variety of clinker kilns are used that differ in the specific energy consumption and CO_2 emission intensity. The specific energy consumption varies from about 3.40 GJ/t for the dry process to about 5.29 GJ/t for the wet process (Madlool et al., 2013). The specific energy consumption of the system is calculated by using the data taken from the factory area for one year (Atmaca and Yumrutaş, 2014). Improvements in energy efficiency are most often achieved by adopting a more efficient technology or production process.

5. Post-combustion CO_2 mitigation options

5.1. Waste heat recovery

A heat recovery unit can recover 50–90% of available thermal energy for space heating, industrial process heating, water heating, makeup air heating, boiler makeup water preheating, industrial drying, industrial cleaning processes, heat pumps, laundries or preheating aspirated air for oil burners.

The waste heat recovered from cement kilns is usually used to dry the raw meals. Depending on the humidity of the raw materials and the cooler technology, additional waste heat is available from the kiln gases (preheater exit gas) and cooler exhaust air. Principally, this heat can be used to dry other materials, such as secondary fuels, or to produce steam or electric power (Ishak and Hashim, 2014). It has been reported that waste heat recovery implemented into the cement manufacturing process can produce 10–30% electric power for a typical cement plant (Karellas et al., 2013), which can reduce the increase in GHG footprint.

5.2. Carbon emissions capture and storage

The International Energy Agency expects that the conventional mitigation measures would only partly fulfil the reduction target for CO_2 emissions, which has been set for 2050 (IEA Cement Technology Roadmap, 2009). This is the reason why carbon emissions capture technologies are being discussed in order to "close this gap". In this context, the capture of CO_2 and its geological storage, often referred to as "carbon capture and storage" (CCS), or its capture and reuse (CCR) in valuable products, is currently being investigated for the cement industry in order to elaborate options, constraints and related cost. For the purpose of CO_2 capture, different categories of technologies are being discussed, but only two seem feasible in the cement clinker production: post-combustion capture as an end-of-pipe solution and the oxyfuel process as an integrated technology. Research activities currently on-going in the field of post-combustion capture include chemical absorption, adsorption, membrane, mineralisation and calcium looping technologies (Schneider, 2015).

Atsonios et al. (2015) presented the integration of calcium looping technology in existing cement plant for CO_2 capture. The study showed that the main advantage of calcium looping technology compared to other CO_2 capturing technologies, which could be applied in the cement industry, is the prospect of reusing purge CaO from calcium looping in cement production as it is chemically compatible with cement raw meal. However, currently the main drawback for the adoption of this novel concept in the cement production process is high equipment cost.

6. Types of alternative raw materials

6.1. Locally available mineral-based materials and construction waste

Typical raw materials in cement production are limestone or chalk ($CaCO_3$), sand (SiO_2), clay (SiO_2, Al_2O_3, and Fe_2O_3), iron ore (Fe_2O_3), and gypsum ($CaSO_4$). Limestone and clay are crushed and blended in a ratio of about 75% limestone to 15% clay, and preheated to drive off water and decompose the limestone into lime and CO_2. The material is then transferred to a rotary kiln, which heats up to 1450 °C, fusing the calcium from limestone and silicon from clay into calcium silicates (Ca_3SiO_5 and Ca_2SiO_4). The resulting clinker is then cooled, ground and combined with 5% gypsum to control setting. To reduce CO_2 emissions and improve cement quality, reactive silica (amorphous silica with average particle size finer than 45 μm) is often used as additive in modern cement production to reduce clinker consumption (Madani Hosseini et al., 2011).

The chemical composition of construction and demolition waste (CDW) is often similar to that of typical raw materials in cement production. In the tests of clinker recipes containing mixtures of typical raw materials with recycled CDW, it was shown that the added recycled aggregates improved the burn ability of the cement raw meal without affecting negatively the clinker properties (Galbenis and Tsimas, 2006).

6.2. Materials derived from agricultural waste

Ash from agricultural wastes, which constitute pozzolanic materials can be used as a replacement for cement. Plant-derived by-products like rice and barley husks, sugar cane, and corn cobs, after they are burned the residue ash contains a certain amount of silica (SiO_2). This residue ash containing silica can be blended with Portland cement, and cement with reduced environmental impact can be obtained. The production of such cement has showed environmental, economic and technical benefits. It reduces clinker consumption and its related energy use and CO_2 emissions. Additionally, plant by-products and other organic residues could substitute fossil fuels, further reducing the CO_2 emissions (Khalil et al., 2014).

6.3. By-products of industrial processes (ashes, iron and steel slag)

Utilisation of supplementary cementitious materials has been found as a suitable measure for the reduction of CO_2 emissions from cement production. Industrial by-products are being used within the cement industry, either as an ingredient for clinker production or as an addition to clinker for cement production. Obviously, cement production based on by-products from other industrial activities must be undertaken with caution. In this sense, the physical and chemical properties of the new cement generated should be comparable to those of standard Portland cement. One goal should be a reduction in the environmental impact of the cement production and use.

One of the by-products most commonly used in cement and concrete manufacture is fly ash from coal-pulverized power plants, which are fine particles collected by electrostatic or mechanical precipitation. Coal power plants also produce bottom ash, which are coarse and glassy particles that fall to the bottom of the furnace and conglomerate. Fly ash represents between 70% and 90% of the coal ashes produced in power plants, while coal bottom ash represents between 10% and 30%. For years now, it has been reported that the addition of fly ash to cement produces properties similar to or even better than Portland cement. One of the main improvements is the lower demand of water and less heat in the hydration process (Menéndez et al., 2014). Despite the existence of these opportunities, a large fraction of the fly ash produced worldwide is still unused and disposed of as waste or stored in landfills. This occurs mostly because cement with larger fractions of fly ash is unable to meet the quality requirements for replacing clinker in cement blends (Vargas and Halog, 2015).

In the cement industry, large quantities of natural gypsum are used in cement production as a set retardant, by adding it to the clinker in a proportion that ranges from 3% to 5%.

The production of titanium dioxide pigments generates the red gypsum as waste. This red gypsum is not treated as dangerous waste, and is neither utilized nor commercialized in any way, however it needs to be appropriately disposed off. A possibility is to use red gypsum instead of natural gypsum in cement industry. It has been reported that cement that has 10% of red gypsum in the mixture with the clinker had comparable characteristics to those of Ordinary Portland Cement which contains 97% clinker and 3% natural gypsum. Meaning that saving on clinker of about 4–5% is possible (Gazquez et al., 2013).

In the oil-drilling industry, large amounts of waste material known as oil-based mud (OBM) are generated at an estimated rate of around 0.37 kg/barrel (159 L) of oil produced. Depending on the geological conditions this material may contain significant quantities of major oxides associated with cement manufacturing: calcium oxide, silicon oxide, and aluminium oxide, along with high oil content. In a study on the use of up to 5% OBM as a raw meal ingredient to produce clinker for cement manufacturing in Oman, it was found that a corresponding percentage reduction of CO_2 emission from clinker burning could be obtained (Abul-Wahab et al., 2016).

For steel manufacturing, calcium oxide or lime (CaO) is added to molten steel at 1650 °C to remove impurities such as silica, magnesium, aluminium, and other oxides. These impurities float to the top and are poured away as slag. This slag can be added to the feed at the end of the kiln as a component of the raw material mix. Because of its lower melting point (1260–1316 °C), the slag does not require additional fuel in the kiln to form clinker with other raw feed components. Moreover, mineralizers already present in the slag help catalyse clinker formation. In addition, the exothermic reaction of converting di-calcium silicate into tri-calcium silicate, which happens when slag is exposed to the high temperature, releases supplementary heat into kiln, resulting in even higher efficiency of the cement manufacturing process (Hasanbeigi et al., 2012). The studies also shown that blast furnace slag and steel slag should preferably be arranged in fine fractions due to their desirable hydration processes and high strength contribution ratios (Zhang et al., 2012).

7. Types of alternative energy sources

7.1. Use of the power (electricity) from renewable energy sources

The power use in cement plants takes place dominantly in raw material preparation, grinding, homogenization and in cement finish grinding. In kilns, the biggest electricity consumers are the drives of rotary kilns. The carbon balance of electricity use is defined by the consumption of electricity (usually expressed as kWh/t cement) and by the CO_2 emission of produced electricity (usually expressed as kg CO_2/MWh). A cement plant can seldom impact the latter, and avoided emission measures are usually concentrated on the efficient use of electricity inside the facility (Kajaste and Hurme, 2015). However, nowadays cement plant operators are increasingly using their electricity provider buying green electricity (Kajaste and Hurme, 2015). This green electricity is produced from wind and solar power plants. In that way cement plant operators are reducing the indirect cement production CO_2 emissions.

7.2. Biomass

Biomass is one of the most extensively used alternative materials in the cement industry because of its diversity and volume. The major restrictions to the use of biomass in cement manufacturing are linked to economic factors, the necessity of pretreatment stages, and the local availability of the resources or the transport costs, which are less restrictive than technical limitations. There is also an issue that biomass burning is producing the other greenhouse gases as NO_X (Čuček et al., 2012b).

Biomass is usually defined as any type of organic material, except those which are catalogued as toxic or hazardous, and those that contain substances such as varnish, paint, or glue. A wide array of different types of biomass is used in combustion or gasification processes, for example, saw dust or wood, straw, agriculture and forest wastes, almond shells, and olive residues. In spite of this wide diversity of biomass types, wood and other waste from agriculture and forest processes are some of the most common types of biomass processed by combustion or gasification.

Biomass additions can replace a portion of the traditional fuel use. Although replacement ratios of approximately 20% are recommended to maintain a stable combustion process and the quality of the clinker, higher values have been used with very satisfactory results.

Calcium (Ca) and potassium (K) are important components in biomass. Because an increase in the potassium oxide (K_2O) content decreases the melting point of the ashes, enhancing agglomeration problems in the combustion chambers, therefore co-combustion of biomass with coal or pet coke, which have lower calcium contents, is recommended. These mixtures produce ashes with a higher melting point and operational problems are thus avoided. Moreover, the alkaline base and chlorine contents are also important, especially at high levels because they can cause deposition, slag fusion, or corrosion problems.

When it comes to the nitrogen content, different biomasses tend to have higher nitrogen content than those in coal or petroleum, and therefore, NOx emissions can be higher. However, taking into account that most of the nitrogen in biomass is converted to ammonia, which promotes the conversion of NOx to gaseous nitrogen, these emissions of NOx can also be reduced (Aranda Usón et al., 2013).

7.3. Meat and bone meal (MBM)

In 1994 European Union banned both the use of meat and bone meal - MBM as cattle feed and the landfilling due to the publicly known mad cow disease. This ban increased the interest in using MBM as fuel in cement industry to ensure that any living organism is thermally destroyed and its energy potential is utilised. Nowadays in France about 45% of the annual production of MBM is burnt in cement plants. The availability of MBM is higher than most of the other alternative fuel commonly used in cement kiln. The feeding rates of MBM in cement kilns vary from country to country. For example, in Spain the limit is 15% of the energy needed in the kilns, but there is no limit in Switzerland. MBM has a lower heating value of 14.47 MJ/kg which is almost half of the coal. Another disadvantage of using MBM in cement industry as fuel is the moisture content which is about 70%. Pre-treatment is required to reduce that, increasing the processing cost. MBM is generally fed in the kiln burner and an additional amount of air may be required if it is used in the calciner. Approximately 5–10% more air is needed for combustion if MBM is fed (Rahman et al., 2015).

7.4. Sewage sludge

Sewage sludge is an organic residue generated by municipalities following secondary and tertiary treatment of wastewater streams. It is used as a soil amendment and fertiliser to improve the yield of selected crops, as well as a fuel in co-combustion with other fuels or types of waste. Sewage sludge is disposed of by land spreading, burial in landfills, and incineration. Sewage sludge can be used in cement production via two different processes: (1) by blending its incinerated ash with Portland cement or (2) by co-combustion of sewage sludge before its addition to Portland cement. Sewage sludge has appreciable quantities of silica, present mainly in minerals such as sand, so both processes could be followed to replace Portland cement. The added advantage of the co-combustion process is that it would allow for some energy recovery from sewage sludge waste. Energy produced during sewage sludge incineration strongly depends on water content of sludge and furnace performance, although its heating value is close to fossil fuel. Consequently, sewage sludge can be considered as raw material and an energy source for cement production, an environmentally friendly alternative to Portland cement – see e.g. Madani Hosseini et al. (2011) later Husillos Rodríguez et al. (2013) and recently Smol et al. (2015).

7.5. Used oils

Waste oil is a hazardous waste that originates from automotive, railway, marine, farm and industrial sources. In European Union approximately 1 million tons of waste oil is used by cement kilns as alternative fuel. Solvent and spent oil from different industries generally have high heating value and those can be used in cement kiln as alternative fuel with minimal processing cost. The range of heating values of solvent and spent oil is between 29 MJ/kg and 36 MJ/kg and the variation occur due to the ratio of different chemical in it. Generally pre-treatment is not required for spent solvent and used oil. Both types of fuel can be fed through the main burner or the calciner using a fuel oil firing system. Un-blended waste oil can also be used to start up the process of the main burner (Rahman et al., 2015).

7.6. Municipal solid waste (MSW) and solid recovered fuel (SRF)

One of the most favourable municipal solid waste - MSW management strategies is thermal treatment or energy recovery to obtain cleaner renewable energy for industries. Among many waste-to-fuel strategies, solid recovered fuel – SRF substitution of fossil fuels is considered as the most advantageous one. SRF is an alternative fuel produced from energy-rich MSW materials diverted from landfills, and it can be used as a substitute energy source in different industries. An industry that is particularly well-suited to the employment of SRF is the cement industry. Furthermore, higher fossil fuel prices are increasingly forcing cement plants to consider the use of SRF for clinker production (Kara, 2012). Finally a significant reduction of GHG emissions can be achieved by replacing conventional fossil fuel with less carbon and resource-intensive SRF (Reza et al., 2013).

Energy recovery of SRF in cement combustion units has some major advantages over regular combustion of SRF in incinerators. Due to the high combustion temperatures inside the cement calciner and rotary kiln, a complete combustion of the waste is ensured. Ash that is produced as a by-product, reacts with the raw material and exits the rotary kiln as clinker, so there is no liquid or solid residue to contend with. This is not the case when the SRF is incinerated or co-fired in utility boilers. The ash from such applications needs to be disposed of in a different way, meaning that there is still a solid residue to contend with (Mikulčić et al., 2015b).

7.7. Used tyres

End life tyre is a waste from automobile industry and generally disposed of in landfills or stockpiles. Landfilling or stockpiling tyres have potential environmental, safety and health hazards like rodent and insect infestation. In mid 1980s tyres became very popular to the cement manufacturers as alternative fuel to cope with the increasing fossil fuel cost. However, the present sharp fall in oil and gas prices is making this and several others options less popular. High carbon content, high heating value of 35.6 MJ/kg and low moisture content make tyre derived fuel (TDF) one of the most used alternative fuels in cement industry around the world. Tyre derived fuel (TDF) costs are significantly lower than natural gas costs and the overall unit cost of tyre derived fuel is even less than the coal. Reinforced wires of tyres can be consumed as a replacement of raw material containing iron when the whole tyre is used as alternative fuel. It has been reported that there exist no significant differences in the chemical composition of the clinker manufactured by using TDF compared to the one manufactured by using fossil fuel. Different form of tyre, from whole to fine grained, can be used in cement kiln as alternative fuel. The fine grained tyre (crumb) can be fed along with powdered coal directly but removal of the steel from tyre to produce crumb is costly (Rahman et al., 2015).

7.8. Plastic waste

Plastic waste is considered as one of the most readily available potential candidates for alternative fuel in cement industry due to its worldwide production and high heating value 29–40 MJ/kg. Plastic waste is available as municipal waste as well as industrial waste. The only concern of using it is the chlorine content which is mainly found in PVC. According to Al-Salem et al., 2009 the accepted particle size for the incineration process is 10 cm × 10 cm × 10 cm and a shredder is needed when larger parts are offered in the kiln. Isolation of materials from plastic waste and retrofitting require additional capital and labour cost. The material preparation can be done in on-site or off-site. However, all those procedures consume energy and are increasing the GHG footprint. Plastic can be conveyed either to the kiln or to the calciner through a belt conveyer (Al-Salem et al., 2010).

7.9. Others

Use of renewable energy resources in cement production has recently attracted worldwide attention. Licht et al. (2012) developed a method for cement production, called Solar Thermal Electrochemical Production of cement or STEP cement, which releases near zero CO_2 emissions. The STEP method uses solar thermal energy instead of the fossil fuel as a heat source. The solar heat is used to melt the limestone, and also provides heat for the electrolysis of limestone. During the electrolysis, depending on the temperature of the reaction, current applied to the limestone ($CaCO_3$) changes the chemical reaction of limestone decomposition. Instead of separating the $CaCO_3$ into lime (CaO) and CO_2, the $CaCO_3$ separates into CaO and some other combination of carbon (C) and oxygen atoms (O). When electrolysed at temperatures below 800 °C, the molten $CaCO_3$ forms CaO, C, and O_2. When electrolysed at temperatures above 800 °C, the products are CaO, CO, and 0.5 O_2. When separated in this way, the carbon and oxygen atoms no longer pose the threat to the environment. As for the CO produced as a by-product at higher temperature, the authors stated that it can be used to produce other fuels, form plastics and other hydrocarbons.

The use of synthetic hydrocarbon fuels in cement production processes is also 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 CO could be produced from sequestered CO_2 or by STEP method and the H_2 could be produced from excess electricity provided by renewable energy resources like wind and solar. From the syngas, hydrocarbon fuels could be produced that could afterwards be used in the production process again. However this type of cement production is still under research (Mikulčić et al., 2013c).

Sebastián González and Flamant (2014) presented a hybrid cement production process that combines the concentrated solar thermal (CST) technology and the cement production process. The study showed that by using CST for the calcination process in the cement production line, CO_2 emissions can be reduced by 40% since no fossil fuel would be used. The study further demonstrated the technical and economic assessment and showed that it is feasible to use concentrated solar thermal technology in the production process of the analysed cement plant.

8. Replacement of cement in concrete or mortar with alternative materials

The rational use of Portland cement is essential for sustainable development of the construction industry. One way to optimize its consumption is the use of supplementary cementitious materials. In addition, the reuse of industrial residues has become a necessity in the current climate, both in terms of industrial efficiency and environmental responsibility (Jacoby and Pelisser, 2015).

Concrete is used worldwide as a building material and is the most consumed substance on Earth after water. The volume of concrete produced globally is approximately 5.3 billion m^3/y, with more than 12 billion t of material used annually. Cement is an essential binding agent in concrete and is key material for satisfying global housing and modern infrastructure needs (Gao et al., 2015).

Several studies concentrated on the possibility to replace cement in concrete or mortar with recycled materials like porcelain polishing residues, glass, recycled tyre rubber, basalt aggregates, ceramic aggregates or other aggregates. Research on alternative binders to Portland cement that reduce the CO_2 emission is progressing, and e.g. the use of alkali-activated binder instead of ground granulated blast-furnace slag cement in concrete or in ordinary Portland cement (OPC)-based concrete reduces the CO_2 emission of concrete by between 55 and 75% (Kajaste and Hurme, 2015).

Waste marble is well usable instead of the usual aggregate in the concrete paving block production (Gencel et al., 2012). The marble waste can be used to improve the physical and mechanical properties of both cement composites and conventional concrete mixtures. The optimum percent of marble sludge that achieve the most appropriate results of physical and mechanical properties in comparison to the control mix is 20% (Mashaly et al., 2015.)

Zeolite is part of a well-defined class of crystalline alumina-silicate minerals. The large quantity of reactive SiO_2 and Al_2O_3 in zeolite chemically combines with the calcium hydroxide produced by the hydration of cement to form additional C—S—H gel and aluminates, resulting in an improved microstructure of hardened cement. Given the growing trend of using natural zeolite in concrete, researchers have begun to investigate the effect of natural zeolite consumption as a supplementary cementitious material on the properties of concrete (Valipour et al., 2014). It was reported that natural zeolite are a suitable replacement for cement in lightweight composites, which could lead to new environmental products such as non-load bearing building materials (Kidalova et al., 2012).

Palm oil fuel ash is a waste material generated in power plant due to burning of palm oil industry waste as a fuel to generate electricity. Annual production of such a massive amount of waste requires a huge disposal field that would be a threat to the environment. Therefore, due to the abundance and high pozzolanic characteristics, palm oil fuel ash has attracted many researchers to evaluate the potential of its use in constructional materials. It was observed that incorporation of palm oil fuel ash in self-compacting concrete enhanced the acid and sulphate resistance, reduced the dry shrinkage and surface water absorption of the self-compacting concrete without an adverse effect in final compressive strength of the products. This observation was reported in a study by Rahman et al. (2014), and in a recent study by Ranjbar et al. (2015).

Majority of these “green concrete” mixes were evaluated from the environmental point of view by means of the Life Cycle Assessment method, and compared with a corresponding conventional concrete mix. The results indicate that the use of the discussed alternative and recycled materials is beneficial in the concrete production industry (Turk et al., 2015).

9. Numerical modelling for cement industry

Numerical modelling and simulations of different thermo-chemical processes inside combustion chambers, represents a valuable method for improving of flow characteristics and the mass and heat transfer within these chambers (Honus and Juchelková, 2014). This method was used for simulating internal combustion engines (Petranović et al., 2015), fuel injection process (Vujanović et al., 2015), selective catalytic reduction for mitigation of NOx from transport sector (Baleta et al., 2015), selective non-catalytic reduction deNOx process for industrial applications (Baleta et al., 2016), fix bed biomass gasification (Mikulandrić et al., 2014), biomass pellet-drop-feed boiler (Gómez et al., 2015), heat transfer on the heat exchangers (Drosatos et al., 2014), large scale utility boilers: under different operating conditions (Al-abbas et al., 2012); under oxy-fuel conditions (Guo et al., 2015); and co-firing of biomass with coal under oxy-fuel conditions (Bhuiyan and Naser, 2015). However considering cement production units, this method was rarely used.

When reviewing peer-reviewed studies published last five years, the Computational Fluid Dynamics (CFD) simulation studies of cement production units, mostly analysed the calciners and just few of them studied the rotary kiln and the cyclone.

9.1. Cyclone

In cement production, cyclone preheating system is used for the heat exchange process between the raw material and the flue gases. The cyclones are located prior to the calciner and the rotary kiln and can have several stages. Inside of a cyclone raw material is heated by moving counter to the flow of the hot flue gases coming from the calciner and the rotary kiln. The particle separation from the gas is done by the centrifugal force acting on the particles directing them to the wall, and separating them from the flow. Additionally due to the gravitational force the particles slide to the lower part of the cyclone and exit the cyclone at its bottom. In contrast to the solid

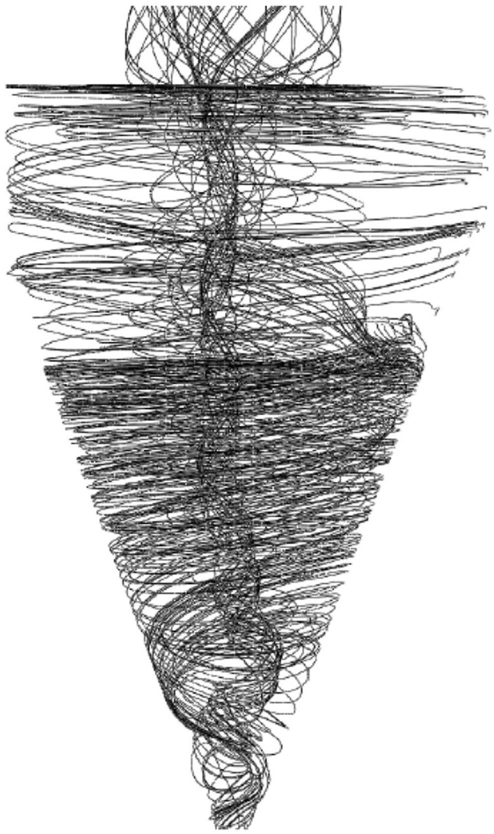

Fig. 6. Flow streamlines inside the cement cyclone (after Mikulčić et al., 2014b).

particles the gas flow has a different behaviour. Firstly the gas swirls downwards in the outer cyclone part, where the separation is done, and then in the lower part of the cyclone where the axial velocity reverses, the gas starts to swirl upwards in the inner cyclone region. This characteristic cyclone gas flow can be observed on Fig. 6.

In a study by Mikulčić et al. (2014b) the reactive multi-phase flow inside of a cement cyclone was investigated. The study showed that gained numerical results, characteristic for cyclones, such as the pressure drop, and particle concentration can thus be used for better understanding of the complex swirled two-phase flow inside the cement cyclone and also for improving the heat exchange phenomena. Wasilewski and Duda (2015) both numerically and experimentally studied geometric configurations of cyclone separators. The study proposed guidelines that enable the design of high-performance cyclones for specific operating conditions. The study also proposed structural changes that may be applied in traditional cyclone dust separators as a solution to the continuously decreasing allowable limits of dust concentration in atmospheric emissions.

9.2. Calciner

Cement calciner is a combustion unit found prior to the rotary kiln. Inside of it combustion of solid fuel, and the endothermic calcination reaction occur (Mikulčić et al., 2012a). With the aim of better understanding of the flow phenomena and heat exchange processes different types of calciners with different operating conditions have been investigated.

In order to improve the limestone degradation rate, Mikulčić et al., 2013a numerically studied the impact of different inlet mass flows and fuel amounts, on the coal burnout rate, limestone decomposition rate, and pollutant emissions. The study showed that CFD is a useful tool for identifying process improvements. In the study by Mikulčić et al. (2014a) different co-firing of biomass with coal conditions in a cement calciner were analysed. The study showed that due to different combustion kinetics of biomass, special attention needs to be given to the complete burnout, in order to avoid undesirable instabilities in the cyclone preheating system. In the recent study by (Mikulčić et al., 2015a) a fully operating in-line cement calciner was numerically investigated. The study showed that the proposed modelling approach can assist in the improvement of the specific local conditions for the calcination process, the reduction of pollutant emissions, and the improvement of the cement calciner's design. The study by Mikulčić (2015) showed that CFD modelling can also be used for estimating the substitution rate of coal, as primary fuel, with solid recovered fuel (SRF), as a secondary fuel. The study showed that there is a maximal allowed coal substitution rate for a stabile cement calciner operation. In Fig. 7 the temperature field inside the calciner for the six calculated cases from this study is shown. In this figure, from the left hand side to the right hand side, the temperature fields for the reference coal case and five SRF co-firing cases are shown. In case (a), where only coal is used as a fuel, the temperature in the near burner region is the highest. When compared to other cases, it can be observed that, from the left hand side to the right hand side, as the thermal share of SRF in the fuel mix is increased, the temperature profile in the near burner region changes. A decrease in the middle of the temperature pick can be observed. This is due to the higher moisture content in the biomass fraction of the SRF than that of coal, meaning that heat is used for drying of SRF.

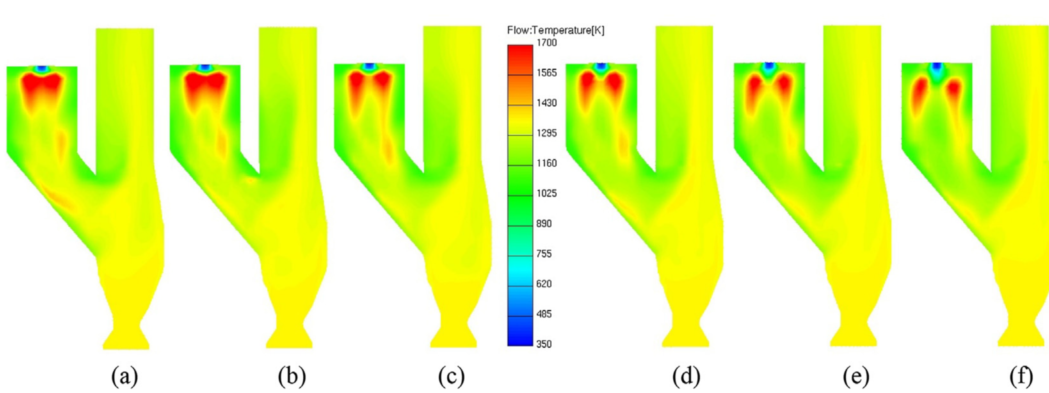

Fig. 7. Temperature fields inside the calciner for the six calculated cases: (a) 100% coal case; (b) 10% SRF co-firing case; (c) 30% SRF co-firing case; (d) 50% SRF co-firing case; (e) 70% SRF co-firing case; (f) 100% SRF co-firing case (Mikulčić, 2015).

All of these studies show that CFD simulation of the complex multiphase flow inside the cement calciners still cannot be considered fully predictive on a quantitative level and that further research is needed.

9.3. Rotary kiln

Production of clinker is the most important process in the cement manufacturing process. The process takes place in a rotary kiln. Over past five year there have been some studies that investigated only the combustion process inside of the rotary kiln.

Ariyaratne et al. (2015) analysed different fuel feeding positions for the meat and bone meal (MBM) combustion in a cement rotary kiln. The study showed the importance of fine fuel grinding, and that MBM particles needed more time to fully combust than coal particles due to the high moisture and slower devolatilisation. Elattar et al. (2014) using the CFD methodology investigated confined non-premixed jet flames in rotary kilns for gaseous fuels in order to understand the flame behaviour and heat transfer. The study presented useful design guidelines and dimensionless correlations that characterize the flame length. Granados et al. (2014) studied the effect of flue gas recirculation (FGR) during coal oxy-fuel combustion in a cement rotary kiln. Simulation results showed that the flame length in the oxy-fuel combustion cases were 30%–65% shorter than that in air combustion with a higher intensity. This could allow shorter kiln designs and might improve high-temperature clinkering reactions during the cement production process. However the study states that more research is needed in this area.

10. Conclusions and possible direction of the future development to a cleaner production of cement

Climate change is one of the serious challenges facing modern society and a reduction of CO_2 emission in cement industry is one of the important measures for achieving climate targets for 2020 and beyond. Even though there is enormous global demand for cement, and the cement production is constantly increasing, the industry is looking for ways to reduce CO_2 emissions from limestone decomposition and fossil fuel combustion.

Several alternative pathways for a more sustainable cement manufacturing including the potential for achieving CO_2 emission reduction have been discussed. These alternative pathways include the reduction of clinker to cement ratio by adding different alternative raw materials, reduction of alternative materials to concrete, the replacement of fossil fuels with alternative fuels, further improvement in the energy efficiency of the existing kiln processes, etc. Available alternative materials and fuels have been summarized and analysed on the ground of advantages, disadvantages, greenhouse gas emissions and environmental impact. This research supports and reinforces the suitability of the use of different waste fuels and materials as alternative energy resources and raw materials in the cement industry.

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