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Modeling the dynamic mechanism between cement CO₂ emissions and clinker quality to realize low-carbon cement

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ABSTRACT

To devise low-carbon cement clinker product, in-depth knowledge about the variation mechanism of cement clinker CO₂ emissions is the prerequisite. The determinants of cement clinker CO₂ emissions in Chinese cement industry have been inquired by several researches recently. However, the impacts and abatement effect of clinker quality on cement clinker CO₂ emissions have not been identified yet. In technological sense, there lacks an integrated quantitative model that can elucidate the dynamic mechanism between cement clinker CO₂ emissions and clinker quality. To fill this gap, this paper devises a CO₂ emission accounting framework integrated with optimization model to explore the dynamic mechanism between cement clinker CO₂ emissions and clinker quality. Based on survey data of 2 typical cement clinker production lines in China, the accounting framework is validated with measured data. Through numerical simulation within this framework, a set of sensitivity analysis is conducted. The results indicate that if the target KH (lime saturation ratio) of both production lines climb from −5% to +5%, the process CO₂ emission factor will give rise to an increase of about 16.4 and 14.8 kg CO₂/t clinker, respectively. With the same variation, the fuel CO₂ emission factors of two production lines will gain 17.2 and 13.7 kg CO₂/t clinker, respectively. Obviously, controlled decrease of clinker quality can reduce CO₂ emission of clinker manufacturing. The results of sensitivity analysis are robust and highlight the remarkable role of clinker quality in both process and fuel emissions. Besides, the integrated framework can provide in-depth investigation into the dynamic mechanism between CO₂ emissions and the other determinants. Using this framework, cement plants or industry can develop optimal clinker quality standards on a quantitative basis to achieve low-carbon clinker.

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1. Introduction

Cement manufacturing has been always ranked in the list of the main sources of carbon emissions (Benhelal et al., 2013; Van Ruijven et al., 2016). According to 'Cement Technology Roadmap to 2050', producing cement has lead the cement industry to generate approximately 5% of global anthropogenic CO₂ emissions (WBCSD and IEA, 2009). Due to mounting demand induced by infrastructure upgrade and urbanization in developing countries (Szabó et al., 2006), especially China, there is a significant growth of cement output; likewise, CO₂ emissions have also risen up (Xu et al., 2013). Cement industry provides an irreplaceable support for the operation of various downstream industries of Chinese economy (Song

et al., 2016) that lead China to be the largest cement producer in the world since 1985 (Ke et al., 2013). CO₂ emissions from China's cement industry have drastically scaled up from 930 Mt in 2005 to 1597 Mt in 2012 while the cement production increased at an average rate of 11% per year during the same period (Zhang et al., 2015b). Since recognizing China's cement industry will inevitably continue to generate large volume of CO₂ emissions, this industry is under close scrutiny recently.

Accurate estimation of cement CO₂ emissions and in-depth interpretation of their variation mechanism are the cornerstone to devise low-carbon cement product. Significant efforts have been implemented to evaluate CO₂ emissions of cement production and to identify the determinants of cement CO₂ emissions. Initially, a regional default emission factor was assumed in estimation of cement emissions. Kim and Worrell (2002) roughly calculated the CO₂ emissions from cement in China. Further, Gregg et al. (2008) presented regional CO₂ emission pattern in China, including cement industry, based on emission data from the Carbon

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Dioxide Information Analysis Center (CDIAC) database. After then, the Cement Sustainability Initiative (CSI) and Intergovernmental Panel on Climate Change (IPCC) default CO₂ emission factors were adopted in several estimations (Ke et al., 2012, 2013; Lei et al., 2011; Liu et al., 2015; Wang et al., 2013; Zhang et al., 2015a,b). However, different data sources are so discrepant with each other (Afsah and Aller, 2010) and there exists significant individual variations of different cement plants instead of a fixed emission factor (Gao et al., 2015). In light of this, Shen et al. (2014) developed a factory-level measurement for cement emission factors primarily using onsite survey and sampling.

On the basis of more and more accurate estimation, further analyses on determinants of cement CO₂ emissions were carried out by the successors. In terms of a comprehensive literature review, CO₂ emissions and energy use at different sections of cement industry have been identified (Ali et al., 2011; Madloul et al., 2011). At the macro level, factors affecting CO₂ emissions in cement industry have been decomposed into energy factor effect, energy structure effect, energy intensity effect, cement production activity effect and clinker production activity effect (Kim and Worrell, 2002; Wang et al., 2013; Xu et al., 2012). In the perspective of plant-level, several scholars proceeded to explore factors and mechanism that affect CO₂ emission of cement production in a single plant (Gao et al., 2015; Gartner, 2004; Popescu et al., 2003). It is argued that the chemistry variations can lead to changes of specific energy consumption (Popescu et al., 2003). Gartner (2004) discussed the inherent characteristics of raw material CO₂ and fuel derived CO₂ in the view of cement manufacturing process. Gao et al. (2015) gave a comprehensive analysis on major determinants of cement CO₂ emissions, which slightly touched upon the clinker quality. On the other hand, efforts have been dedicated to designing low-carbon cement. Incorporation of industrial wastes, such as, waste glasses (Shi and Zheng, 2007), recycled concrete waste (Gastaldi et al., 2015; Oh et al., 2014), oil-based mud (Abdul-Wahab et al., 2016; Bernardo et al., 2007), arc furnace steel slag (Bernardo et al., 2007; Iacobescu et al., 2011) and contaminated soil (Navia et al., 2006) into the clinker process, are found to be feasible to reduce the CO₂ emissions of cement manufacturing.

These above studies have, to a certain extent, contributed to gaining the insight into determinants of cement CO₂ emissions and design of novel low-carbon cement. However, we note that the influence of clinker quality on cement CO₂ emissions is neglected by previous researches. This is because there lack a quantitative model to investigate the dynamic mechanism between CO₂ emissions and clinker quality. Therefore, the aims of this work are as follows:

- An integrated accounting framework of CO₂ emission factors of cement industry incorporating clinker quality and a proportioning model is established to reveal the dynamic mechanism between CO₂ emission and clinker quality.
- This framework is to be validated by empirical data collected from 2 cement production lines.
- By operating the simulation of this accounting framework, the abatement effect of clinker quality on CO₂ emissions are quantitatively evaluated through sensitivity analysis.

2. An overview of clinker manufacturing

Because the CO₂ emissions derive primarily from the production of clinker (Josa et al., 2004), the scope of this study focuses on the clinker manufacturing. A typical new suspension preheater (NSP) process consists of crushing, pre-homogenization, raw material grinding, filter, preheating and pre-calcining, pyro-processing, cooling, grinding, storage and packing or shipping (Fig. 1). In the

pre-homogenization unit, a proportioning procedure of different raw materials is indispensable. The proportioning procedure is an important process because the blending procedure will affect clinker quality (Li et al., 2012).

The production of clinker which is an intermediate product is the most important component of Portland cement (Boesch et al., 2009). Most chemical reactions are occurred in the process of clinker manufacturing.

Chemically, the main compositions of clinker are calcium oxide (CaO) and silica (SiO₂), lesser amount of aluminum oxide (Al₂O₃) and iron oxide (Fe₂O₃) (Van Oss and Padovani, 2002). Given the dominance of CaO in clinker, raw materials must contain a high percentage of this oxide. In practical terms, limestone is the major raw material used in production of clinker (Gao et al., 2015). During the calcination process, lime decomposes into lime and CO₂ as a by-product. CO₂ is emitted from the thermal decomposition of calcium carbonate (CaCO₃) and magnesium carbonate (MgCO₃), which can be expressed by following stoichiometric formulas:

As the kiln temperature rises, lime reacts with silicate, aluminate and ferrite in sintering process to form clinker minerals (CSI, 2011). The clinker production can be expressed by a series of chemical formulas:

3. CO₂ emission accounting framework integrating clinker quality

3.1. The integrated CO₂ emission accounting framework

The purpose of this emission accounting framework is to establish the casual relationship between CO₂ emission and clinker quality. For the purpose to analyze how the clinker quality influences CO₂ emission, we propose an accounting framework of CO₂ emission factor incorporating clinker craft parameters and a proportioning model. As shown in Fig. 2, the main steps of the integrated CO₂ emissions accounting framework of cement production are as follows.

- (1) The specific coal consumption is calculated by the heat efficiency of the clinker kiln and clinker quality objectives on the basis of thermochemical equilibrium (GB/T 26281-2010, 2011);
- (2) The proportion of coal ash mixed into the clinker is deduced by the specific coal consumption in step (1);
- (3) The CO₂ emission factor of kiln fuel is derived from the specific coal consumption (Shen et al., 2014);

Fig. 1. Processes and CO₂ sources of a typical new suspension preheater (NSP) cement production line. Note: This figure is modified from Ishak and Hashim (2015)'s study.

Fig. 2. Integrated CO₂ emissions accounting framework.

- (4) Based on the clinker quality objectives, the proportion of coal ash added into clinker, and chemical content of raw feeds, a proportioning model provides optimal ratio of raw feeds;
- (5) Subsequently, the process CO₂ emission factor is obtained from the ratio of carbonate in raw material (Shen et al., 2014).

To better understand the relationship between CO₂ emissions and clinker quality, we provide a brief introduction to clinker quality control indicators and specific fuel consumption of clinker production in the following Sections 3.2 and 3.3.

3.2. Clinker quality control indicators

Clinker quality largely depends on the content of the hydraulic materials in clinker. A typical Portland cement clinker contains four principal minerals: Alite (3CaO·SiO₂, shortened to C₃S), Belite (2CaO·SiO₂, shortened to C₂S), Aluminate (3CaO·Al₂O₃, shortened to C₃A), Ferrite (4CaO·Al₂O₃·Fe₂O₃, shortened to C₄AF). These four minerals provide functional properties of clinker, of which C₃S and C₂S account for approximately 75% in by weight. By hydration, a clinker with higher C₃S will exhibit greater early strength performance but reduced rate of mechanical strength development at

later age of hydration. Although C₃A and C₄AF can provide strength in certain degree, the main function of them is to lower the sintering temperature and to facilitate the formation of C₃S.

Commonly, lime saturation ratio (KH), silicic ratio (SM) and aluminum ratio (IM) are control indicators of clinker quality widely adopted by cement plants in China (Zhou and Peng, 2009).

KH: As shown in Eq. (1), the KH is a ratio of CaO to other three main oxides, which controls the ratio of C₃S to C₂S in the clinker.

$$KH = \frac{Clinker_C - 1.65 \times Clinker_A - 0.35 \times Clinker_F}{2.8 \times Clinker_S} \quad (1)$$

- $Clinker_C$, $Clinker_S$, $Clinker_A$ and $Clinker_F$ denote the content of CaO, SiO₂, Al₂O₃ and Fe₂O₃ in clinker (wt%), respectively.

SM: As defined in Eq. (2), the SM is a ratio of SiO₂ to the sum of Al₂O₃ and Fe₂O₃, which determine the proportion of silicate in clinker. A higher SM refers to more silicate in clinker. In other word, an increase of SM will also improve the strength of clinker, i.e., clinker quality.

$$SM = \frac{Clinker_S}{Clinker_A + Clinker_F} \quad (2)$$

IM: As presented in Eq. (3), the IM is a ratio of Al_2O_3 to Fe_2O_3 . The IM determines the potential relative proportions of C_3A and C_4AF , which has significant effect on formation of C_3S in sintering process. The content of C_3A will proportionally increase with the augment of IM, whereupon the formation of C_3S is unfavorable under the condition of excessive IM.

$$IM = \frac{\text{Clinker}_A}{\text{Clinker}_F} \quad (3)$$

The above three clinker quality indicators fluctuate within a certain range according to desired functional properties of different cement. In China, the conventional aberrations of KH, SM and IM are aimed at 0.85–0.96, 1.7–2.7 and 0.9–1.7. These values are fundamentals for cement production which can be found in the production guideline in a cement plant (Zhou and Peng, 2009).

3.3. Specified fuel consumption

Generally speaking, the sintered temperature of clinker must maintain at the range of 1300–1450°C. To sustain the rated temperature of burning zone, a certain quantity of fuel need to be fed into the calcining kiln by several times. Theoretically, producing one ton of clinker requires at least 1.6 GJ heats (Liu et al., 1995). However, the energy efficiency of entire cement system is about 50% due to the heat loss (Engin and Ari, 2005). According to the thermochemical equilibrium, the specific fuel consumption of clinker is presented in Eqs. (4) and (5) (GB/T 26281–2010, 2011).

$$Q_f = \frac{Q_{sh}}{\eta} \quad (4)$$

$$Q_{sh} = 17.19 \times \text{Clinker}_A + 27.1 \times \text{Clinker}_M + 32.01 \times \text{Clinker}_C - 21.4 \times \text{Clinker}_S - 2.47 \times \text{Clinker}_F \quad (5)$$

- Q_f denotes the actual heat consumption (kJ/kg clinker); Q_{sh} denotes heat of clinker formation (kJ/kg clinker); η denotes the heat efficiency of rotary kiln system (%); Clinker_M refers to the content of MgO in clinker.
- Note: This equation is a conventional indicator for evaluating the heat efficiency of cement kiln (Engin and Ari, 2005). It refers to the reaction heat required to form the clinker without any heat loss based on the chemical composition in it.

For availability reason, the chemical ingredients in clinker may be partly contributed by the kiln fuel. In China, coal is the traditional fuel used in cement industry (Gao et al., 2015). After combustion, the coal ash, which contains similar oxides as raw material, is mixed into the clinker. The coal ash added in clinker is calculated according to Eq. (6):

$$\text{Ash}_{\text{clinker}} = \frac{Q_f}{Q_{\text{net}}} \times \text{Ash}_{\text{coal}} \quad (6)$$

- $\text{Ash}_{\text{clinker}}$ refers to the coal ash added in clinker (wt%); Q_f denotes the actual heat consumption (kJ/kg clinker); Q_{net} refers to net calorific value of coal (kJ/kg coal); Ash_{coal} refers to ash content of coal (wt%).

3.4. Proportioning model of raw material

In order to produce a qualified clinker, raw materials should be reasonably mixed. Therefore, a blending process is required before

raw feeds are transported into the burning kiln. In blending process, appropriate ingredient ratio should adapt to the oxides in raw materials and guarantee the KH, SM and IM of clinker in permissible scope (Li et al., 2012). According to Sections 3.1 and 3.3, chemical composition from raw material and coal ash codetermines these three critical craft parameters of clinker. In other words, clinker quality is the constraint condition on the ratios of raw materials and coal ash. Calculation for raw material proportion can be regarded as a programming optimization problem. Based on the mass balance and energy balance, a nonlinear optimization model of raw material mix is built, which includes multiple objective functions (Eq. (7)) and constraint equations (Eq. (8)).

Minimize : $KH_{\text{calculation}} =$

$$\frac{\text{Clinker}_C - 1.65 \times \text{Clinker}_A - 0.35 \times \text{Clinker}_F}{2.8 \times \text{Clinker}_S},$$

$$SM_{\text{calculation}} = \frac{\text{Clinker}_S}{\text{Clinker}_A + \text{Clinker}_F}, IM_{\text{calculation}} = \frac{\text{Clinker}_A}{\text{Clinker}_F} \quad (7)$$

• Subject to:

$$\begin{cases} \Sigma r_i = 100\% (8) \\ KH_{\text{calculation}} = \frac{\text{Clinker}_C - 1.65 \times \text{Clinker}_A - 0.35 \times \text{Clinker}_F}{2.8 \times \text{Clinker}_S} \geq KH_{\text{target}} \\ SH_{\text{calculation}} = \frac{\text{Clinker}_S}{\text{Clinker}_A + \text{Clinker}_F} \geq SM_{\text{target}} \\ IM_{\text{calculation}} = \frac{\text{Clinker}_A}{\text{Clinker}_F} \geq IM_{\text{target}} \end{cases} \quad (8)$$

- $KH_{\text{calculation}}$, $SM_{\text{calculation}}$ and $IM_{\text{calculation}}$ denote the calculated value of KH, SM and IM, respectively;
- KH_{target} , SM_{target} and IM_{target} denote the target value of KH, SM and IM;
- r_i represents ingredient ratio of raw material i (%);

The following equations are the illustration for Eqs. (7) and (8), which are shown in Eqs. (9)–(12). These equations are based on the chemical equilibrium illustrated in Section 3.1 and Eq. (13) is derived from Eqs. (4)–(6).

$$\begin{aligned} \text{Clinker}_C &= \text{Ash}_{\text{clinker}} \times \text{Ash}_C + (1 - \text{Ash}_{\text{clinker}}) \\ &\times \frac{\Sigma (r_i \times RC_i)}{1 - \Sigma (r_i \times L_i)} (i = 1, 2, 3 \dots) \end{aligned} \quad (9)$$

$$\begin{aligned} \text{Clinker}_S &= \text{Ash}_{\text{clinker}} \times \text{Ash}_S + (1 - \text{Ash}_{\text{clinker}}) \\ &\times \frac{\Sigma (r_i \times RS_i)}{1 - \Sigma (r_i \times L_i)} (i = 1, 2, 3 \dots) \end{aligned} \quad (10)$$

$$\begin{aligned} \text{Clinker}_A &= \text{Ash}_{\text{clinker}} \times \text{Ash}_A + (1 - \text{Ash}_{\text{clinker}}) \\ &\times \frac{\Sigma (r_i \times RA_i)}{1 - \Sigma (r_i \times L_i)} (i = 1, 2, 3 \dots) \end{aligned} \quad (11)$$

$$\begin{aligned} \text{Clinker}_F &= \text{Ash}_{\text{clinker}} \times \text{Ash}_F + (1 - \text{Ash}_{\text{clinker}}) \\ &\times \frac{\Sigma (r_i \times RF_i)}{1 - \Sigma (r_i \times L_i)} (i = 1, 2, 3 \dots) \end{aligned} \quad (12)$$

$$\text{Ash}_{\text{clinker}} = \frac{17.19 \times \text{Clinker}_A + 27.1 \times \text{Clinker}_M + 32.01 \times \text{Clinker}_C - 21.4 \times \text{Clinker}_S - 2.47 \times \text{Clinker}_F}{Q_{\text{net}} \times \eta} \times \text{Ash}_{\text{coal}} \quad (13)$$

- Ash_C , Ash_S , Ash_A and Ash_F refer to the content of CaO, SiO₂, Al₂O₃ and Fe₂O₃ in coal ash (wt%), respectively;
- RC_i , RS_i , RA_i and RF_i refer to the content of CaO, SiO₂, Al₂O₃ and Fe₂O₃ in raw material i (wt%);
- r_i represents ingredient ratio of raw material i (%);
- L_i represents loss on ignition of raw material i (wt%);
- $i = 1, 2, 3, \dots$ denotes raw material species.

In this model, the calculated KH, SM and IM, which are derived from the results of optimal ingredient ratio, should approach the target standard of clinker quality as far as possible while these three indicators are subject to constraint condition.

3.5. CO₂ emission accounting method

It is widely accepted that CO₂ emissions of cement production can be divided into three types, i.e., process-related emission, combustion-related emission and electricity-related emission (Ali et al., 2011). With respect to emission source, the main direct CO₂ emissions are result of the chemical reactions in the clinker kiln and the fuel combustion during clinker production (Josa et al., 2004; Worrell et al., 2001). The CO₂ emission associated with the electricity consumed is considered indirectly (Ke et al., 2013). Most of the electricity consumed in cement production is purchased from external grid and used to crush raw material, to mash raw material mix, to operate fans and coolers and to grind clinker with additives to produce cement product (Gao et al., 2015; Rosković and Bjegović, 2005). In the cement production with dry process, only about 35% of electricity is consumed at the clinker grinding stage (Ali et al., 2011). The behavior of the clinker during clinker grinding stage is directly related to the texture and crystal size (Tsvilis and Kakali, 1997). It is verified by experimental samples that the specified fineness is the primary influential factor of the clinker grinding electricity requirement while the clinker quality has no significant impact on the clinker grinding electricity requirement (Tokuyay, 1999). In addition, the trace elements have certain effect on clinker grinding electricity consumption (Opoczky and Gavel, 2004), which are not related to the clinker quality. Thus, in the present paper, we confine the research scope on the theoretical process-related emission and combustion-related emission.

Efforts to account CO₂ emissions of Chinese cement manufacturing have been made by many scholars and organizations (CMBA, 2011; CSI, 2011; He, 2009; IPCC, 2006; Shen et al., 2014; Wang, 2009). Basically, all of these methods comply with material conservation and energy balance. Based on one of recent proposed calculation method (Shen et al., 2014), we calculate process-related emission factor and combustion-related emission, respectively.

3.5.1. Combustion-related emission

Almost all fuels are consumed in the rotary kiln which is heart and the most energy consuming part of a conventional clinker production line (Atmaca and Yumrutaş, 2014). The CO₂ emission factor of fossil fuel combustion is calculated based on the specific fuel used in clinker production and the fuel CO₂ emission factor, which is expressed in Eq. (14):

$$EF_f = Q_f \times F \times 1000 \quad (14)$$

- EF_f denotes the fuel-related emission factor (kg CO₂/t clinker);
- F denotes the default emission factor of fuel (kg CO₂/kJ).
- Note: IPCC (2006) provides a set of default emission factor of different fuel type. Nevertheless, the emission factor suggested by IPCC is not in accordance with the fuel used in Chinese cement plant (Zhao and Wei, 2013). A cement plant purchases coal from different suppliers thus uses coal being mixed with other types of coal. Here, we adopt a measure emission factor

of 93832.32 kg CO₂/TJ (1 TJ = 10⁹ kJ) suggested by Zhao and Wei (2013), which is more practical for Chinese clinker production.

It is noted that the diesel is not incorporated in the fuel emission factor. Generally, diesel is used for firing up the kiln and is not involved in the clinkerization reaction other than coal. Thereupon, the diesel-related emission is not affected by clinker quality and not discussed in this work.

3.5.2. Process-related emission

Unlike fossil fuel emissions, industrial CO₂ emissions are process-related CO₂ emissions resulting from the calcination of carbonate materials, which account for more than 50% of total emissions (IPCC, 2006; Shen et al., 2014). The process-related CO₂ emissions mainly come from precalcining and pyro-processing of carbonate in clinker production stage (Shen et al., 2014). The process-related CO₂ emission can be calculated in two methods: raw material method and clinker method.

The former method using CO₂ content of carbonate-containing raw material and the amount of raw material consumed for per ton clinker to deduce the process CO₂ emission of clinker (Eq. (15)):

$$EF_{pro}^r = \frac{(1 - Ash_{clinker}) \times r_{carbonate} \times (C_{carbonate} \times (44/56) + M_{carbonate} \times (44/40))}{1 - \sum (r_i \times L_i)} \times 1000 \quad (15)$$

- EF_{pro}^r denotes the process-related CO₂ emission factor using raw material method (kg CO₂/t clinker);
- $Ash_{clinker}$ refers to the coal ash added in clinker (wt%);
- $r_{carbonate}$ denotes share of carbonate in raw materials (%);
- r_i represents ingredient ratio of raw material i (%);
- L_i represents loss on ignition of raw material i (wt%);
- $C_{carbonate}$ and $M_{carbonate}$ refer to the content of CaO and MgO in the carbonate (wt%), respectively.

The clinker method, propose by IPCC Tier 2, is based on content of CaO and MgO in the clinker (Ke et al., 2013):

$$EF_{pro}^c = (Clinker_C \times \frac{44}{56} + Clinker_M \times \frac{44}{40}) \times 1000 \quad (16)$$

- EF_{pro}^c denotes the process-related CO₂ emission factor using clinker method (kg CO₂/t clinker);
- $Clinker_C$ and $Clinker_M$ refer to the content of CaO and MgO in clinker (wt%), respectively.

4. Data collection of case cement plants

We select two typical cement production lines with different capacity in Liaoning province as cases to simulate the dynamics mechanism between CO₂ emissions and clinker quality. To ensure the consistency and completeness, year-round data of assay and production are collected from the historic accounting documents of these production lines. Table 1 presents the summarized production status and baselines for target clinker quality of the surveyed plants. NSP technique is adopted by these manufacturers of which the ordinary Portland cement is the major product. The main carbonate provider of these four production lines is limestone. The type of coal conventionally used in these four lines is anthracite or bitumite. Clay or sandstone is used as the siliceous material as well as coal gangue. Some industrial byproducts from adjacent factories or mines (e.g., iron tailings, converter slag and sulfate slag) are recycled as rectifier in raw material mix and can be used as substitute for limestone (Song et al., 2016).

Table 1
Production status and baselines for target clinker quality of two surveyed plants.

Plant name	Production line 1 (PL1)	Production line 2 (PL2)
Date of data	2013	2012
Clinker production capacity (t/day)	2000	5000
Annual clinker output (kt/year)	360	1376
Heat efficiency of rotary kiln system (%)	45.69	55.06
Specific coal consumption (kJ/kg clinker)	3736.01	3173.41
Mean annual lower heating value of the coal (kJ/kg)	20,208.76	25,599.35
Baseline of target KH	0.895234	0.898436
Baseline of target SM	2.777166	2.509841
Baseline of target IM	0.954748	1.295553

Table 2
Process-related emission and fuel-related emission of two cases (unit: kg CO₂/t clinker).

	EF_{pro}^r	EF_{pro}^c	EF_f
Production line 1	493.409	535.186	350.558
Production line 2	533.297	537.980	297.768

Collected data are documented in a spreadsheet in which the calculation formulas of the accounting framework are preset. Detail assay data regarding clinker production are registered in the spreadsheet, as well as energy use. The assay data provide the chemical composition regarding each raw material for clinker production and that of the coal ash while the energy use data include the heat efficiency of rotary kiln system, the specific coal consumption and the mean annual lower heating value of coal. Additionally, to validate the simulated results, the actual craft parameters of clinker product and chemical composition of raw material mix are collected. The detailed information is presented in [Appendix A](#).

5. Simulation results

To analyze the dynamic mechanism, we design a set of sensitivity analysis based on the simulation results of CO₂ emission accounting model and set the target clinker quality in [Table 1](#) as baseline scenario. Sensitivity analysis of a model output aims to identify the influence direction and quantify the relative importance of each input model parameter ([Homma and Saltelli, 1996](#)). Assuming that a variation from the basic scenario is $\pm 5\%$, we try to figure out the influence of clinker quality.

5.1. Simulation results of proportioning model for baseline scenario

Based on the proportioning model and information from [Appendix](#), to achieve the baseline target value of KH, SM and IM in [Table 1](#), the ingredients ratio of each raw material is computed through programming solution. After that, process-related and fuel-related CO₂ emission of clinker is calculated simultaneously. Results of the two case studies are shown in [Table 2](#). Compared to clinker method, the gap between two process emission calculating methods in PL1 is larger than PL2. This is because the CaO and MgO in clinker partially come from the alternative raw material and the clinker method will exaggerate CO₂ emission from carbonate decomposition ([Gao et al., 2015](#)). Hence, the process-related emission derived from raw material method is adopted by this study.

Furthermore, simulation results of raw material and clinker are compared with measured data to validate whether the simulation results are within an acceptable degree of uncertainty. The model mirrors actual raw material and clinker with an error of around 5%. Errors of raw material mainly come from grinding loss. In a

dry type clinker production, there exists mass loss in the raw material preparation process ([Huntzinger and Eatmon, 2009](#)). In another word, raw material grinding releases few particulate emissions and causes deviation between calculated value and factual value. Respecting error of clinker, it is mainly attributed to incomplete reaction. The ignition losses of clinker in two cases are 0.26% and 0.16%, respectively. Theoretically, the input raw material and output clinker should reach equilibrium in the rotary kiln and there is no ignition loss in clinker. In practical, however, perfect calcination and clinkerization are almost unattainable. Since the variation of the simulation results is within a reasonable range, we can further explore the relationship between clinker quality parameters and CO₂ emission based on this simulation.

5.2. Sensitivity analysis of dynamic relationship between clinker quality and CO₂ emissions

On the one hand, process CO₂ emission is determined by CaO and MgO content in clinker coming from carbonate while fuel CO₂ emission is partially determined by CaO and MgO content in clinker according to Eqs. (4) and (5). On the other hand, the oxide compositions in clinker depend on the clinker quality control indicators. Therefore, there exists a dynamic relationship between clinker quality and these CO₂ emission factors.

5.2.1. Clinker quality and process CO₂ emission factor

Sensitivity analysis for impact of different clinker quality control indicators on process CO₂ emission factor are given in [Figs. 3 and 4](#). In production line 1, if the target KH increases from -5% to $+5\%$, the process CO₂ emission factor will present an uptrend from 484.988 kg CO₂/t clinker to 501.347 kg CO₂/t clinker. Consistent tendency is displayed in the two other indicators as well as in production line 2. Within the variation of -5% to $+5\%$, each indicators show a positive influence on process CO₂ emission factor with a correlation coefficient greater than 0.999.

Among the three indicators, the most significant indicator that contributes to process CO₂ emission factor is KH, followed by IM and SM. The increase of KH directly levels up the CaO content in clinker, thereby claiming more calcium oxide provider. Thus, the proportion of carbonate is raised and the process CO₂ emission factor is increased as well. Irrespective of the efficiency of the calcination process, every ton of C₃S produces 579 kg of CO₂ ([Cassagnabère et al., 2009](#)), which release more CO₂ than C₂S. In addition, the increase of IM and SM will change the relative ratio of silicon oxide, aluminum oxide and ferrite oxide, which require more calcium oxide to satisfy the chemical equilibrium.

5.2.2. Clinker quality and fuel CO₂ emission factor

We apply the same sensitivity analysis on fuel CO₂ emission factor, of which the results are presented in [Figs. 5 and 6](#). In both of production lines, the KH value exerts positive effect on fuel CO₂ emission factor. In production line 1, every 1% increase of target KH will lead to an increase of 1.72111F¹ kg CO₂/t clinker for fuel emission, correspondingly. The regression result of production line 2 is akin to that of production line 1. The influence of SM and IM is rather minor compared to KH, though SM has a negative impact on fuel emission factor.

As mentioned in [Section 3.2](#), it is obvious that an increase in KH leads automatically to an increase in C₃S content and a decrease in C₂S content. In clinker production, the formation of C₃S occurs at higher temperature than the formation of C₂S ([Popescu et al., 2003](#)). Besides, higher KH means the augment of carbonate proportion in

¹ $1.7211 = 192.2 \times 0.895234 \times 1\%$.

Fig. 3. Sensitivity analysis of process CO₂ emission factor in production line 1. Fig. 4. Sensitivity analysis of process CO₂ emission in production line 2. Fig. 5. Sensitivity analysis of fuel CO₂ emission in production line 1.

raw material mix thus more heat is needed to de-carbonate the carbonate (Damtoft et al., 2008). Overall, KH has remarkable influence on fuel CO₂ emission.

5.2.3. Other factors affecting CO₂ emission factors

To figure out the other determinants of clinker CO₂ emission factor, we perform the comparative simulation by setting same clinker control indicators for the two cases. KH, SM and IM of production line 1 are set as same as that of production line 2. The simulation results in Table 3 indicate that the varying heat efficiency and raw

Table 3

Comparative simulation of two production lines with same clinker quality.

Production line	PL1	PL2
EF_{pro} (kg CO ₂ /t clinker)	507.825	533.297
EF_f (kg CO ₂ /t clinker)	357.177	297.768
Heat efficiency of rotary kiln system (%)	45.69	55.06
Alternative raw material added	converter slag	not added
SiO ₂ content in raw material mix (%)	13.02	13.63
MgO content in raw material mix (%)	1.79	1.91

Fig. 6. Sensitivity analysis of fuel CO₂ emission in production line 2.

material prescription will also produce significant discrepancy in both of process emission and combustion emission.

The influence of heat efficiency and raw material prescription are evaluated by analogous sensitivity analysis. As presented in Fig. 7, the influential coefficients of the heat efficiency of rotary kiln to fuel CO₂ emission factor are -767.9 and -541.7, respectively. With every 1% improvement of heat efficiency of rotary kiln, the fuel CO₂ emission factor will respectively decrease 3.5112 kg CO₂/t clinker and 2.98948 kg CO₂/t clinker in production line 1 and production line 2. In regard to process CO₂ emission, the promotion of heat efficiency will slightly elevate the process CO₂ emission factor. Reduced coal use means less coal ash added in clinker thus requiring more CaO from raw material. Nevertheless, the impact of heat efficiency on process CO₂ emission is negligibly low. This suggests that the decrease of fuel emission will offset the increase of process emission. China has raised investment in more efficient technologies for cement industry (Oggioni et al., 2011), namely, deployment of current state-of-the-art technologies in new cement plants or the retrofitting of energy-efficiency equipment in old cement plants (Xu et al., 2014). Overall, improvement of heat efficiency will reduce the CO₂ emission of clinker. Chinese cement industry has energy-related CO₂ emission reduction potential of at least 1 Giga tones in a realistic continuous improvement pace (Ke et al., 2012).

According to Table 3, the SiO₂ content and MgO content of production line 2 is higher than that of production line 1 while the raw material substitution of production line 2 is lower than that of production line 1. The effect of dominant chemical composition in each corrective is examined. As presented in Fig. 8, the SiO₂ content of siliceous and MgO content of carbonate show positive influence on process CO₂ emission factor. If the SiO₂ content of siliceous material, CaO content of alternative material and MgO content of carbonate in production line 1 rise or fall to the same level in production line 2, the process CO₂ emission of the former will increase 15.755, 8.814 and 4.410 kg CO₂/t clinker, correspondingly. Subject to the feedstock sources, it is unrealistic to change the siliceous material and limestone without taking account of economic cost. Nevertheless, the use of alternative raw material is feasible (Buruberry et al., 2015; Chen et al., 2010; Kajaste and Hurme, 2016; Shih et al., 2005; Zhang et al., 2014; Zhang and Mabee, 2016) and can reduce the process CO₂ emission (Gäbel and Tillman, 2005). Efficient utilization of industrial wastes in cement manufacturing plays a significant role in reducing the CO₂ emissions of cement industry (Zhang et al., 2013). When concomitant reductions in fuel consumption are also considered, total CO₂ reductions can in theory be as high as 25% (Damtoft et al., 2008).

6. Conclusions

Cement clinker production is a complicated chemical reaction process of which the raw material components may vary from different production lines. To identify the dynamic mechanism between clinker CO₂ emission and various factors, we construct an integrated CO₂ emission accounting framework for cement clinker combined CO₂ emission with clinker quality. The accounting framework provides insights into sensitivities of CO₂ emissions to clinker quality. Not only that, it is a delicate model to simulate the influence of any factors on CO₂ emission factors.

We first give an overview of production process and chemical fundamentals of clinker. Based on the chemical equilibrium and energy balance, we propose a nonlinear optimization model of proportioning raw material, and then embed the CO₂ emission accounting method into this model. To validate the model, we operate the simulation of the nonlinear optimization model with survey data from 2 case clinker production lines. Whereafter, a series of sensitivity analysis is implemented to examine the correlation between CO₂ emission factors and clinker quality as well as other factors. Although sensitivity analysis cannot provide linear formulas of the relationship between clinker quality and CO₂ emission factors, it still gives insight into the interaction mechanism of them.

As a result, each clinker quality control indicators shows positive effect on process CO₂ emission. Except for silicic ratio (SM), the other two indicators have significant positive impact on fuel CO₂ emission. The above results imply that controlled reduction in the clinker quality parametric value can reduce CO₂ emission of clinker manufacturing. Also, there is a positive linear correlation between the thermal efficiency of rotary kiln and the cement clinker CO₂ emission. It is verified that improvement of energy efficiency for rotary kiln can reduce the overall CO₂ emission during clinker production. Additionally, the presence of alternative raw material can mitigate the decarboxylation reaction in clinker production while the decrease of SiO₂ content in siliceous material and MgO content in carbonate will decrease the process CO₂ emission.

The production of clinker is the most energy and CO₂ intensive stage in the production of cement (Deja et al., 2010; Kim and Worrell, 2002). This work has investigated the influence of clinker quality on CO₂ emissions. In turn, the integrated accounting framework proposed in this work can be used as a decision-making tool for cement production. Based on the simulation of this tool, managers of cement plants and policy makers of cement industry could make decisions on developing clinker quality standards and raw material mix portfolio to achieve low-CO₂ clinker. Finally, it is worth to mention the limitation of this work. Due to the lack of

Fig. 7. Sensitivity analysis of heat efficiency of rotary kiln of line 1 (a) and line 2 (b).

Fig. 8. Sensitivity analysis of SiO₂ content of siliceous material, CaO content of alternative material and MgO content of carbonate.

sufficient data, our simulation just provides a theoretical framework and a preliminary empirical study on clinker CO₂ emission factor. Undoubtedly, a more extensive survey is required for the future research on more detailed analysis on determinants of CO₂ emission factors.

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Appendix A.

Table A1

Chemical composition of raw materials and coal ash for production line 1 (unit: %).

Name	Loss	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO
Raw material I	41.85 (0.76)	3.60 (1.32)	0.75 (0.26)	0.41 (0.14)	50.69 (1.64)	1.66 (0.73)
Raw material II	19.17 (2.63)	46.31 (3.25)	19.49 (0.77)	10.01 (1.29)	1.29 (0.34)	0.94 (0.08)
Raw material III	5.07 (2.10)	18.75 (1.23)	4.46 (0.51)	19.30 (1.06)	42.35 (1.62)	5.66 (0.48)
Raw material IV	2.13 (0.23)	73.32 (0.75)	6.70 (0.64)	8.53 (0.74)	2.79 (0.24)	2.40 (0.28)
Ash content of coal	20.98 (0.78)					
Coal ash		58.65 (1.30)	23.01 (1.12)	7.84 (0.80)	2.54 (0.40)	1.90 (0.18)
Raw material mix (measured)	34.46 (0.42)	14.18 (0.21)	1.92 (0.12)	2.63 (0.32)	43.34 (0.46)	2.32 (0.13)
Clinker (measured)	0.26 (0.04)	22.20 (0.28)	3.90 (0.18)	4.09 (0.44)	63.51 (0.44)	3.44 (0.37)

Note: Standard deviation is displayed in the parentheses.

Table A2

Chemical composition of raw materials and coal ash for production line 2 (Unit: %).

Name	Loss	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO
Raw material I	38.19 (1.74)	9.34 (2.88)	1.98 (0.68)	1.07 (0.31)	46.25 (1.99)	2.00 (0.44)
Raw material II	8.60 (0.52)	57.58 (2.53)	19.77 (1.30)	8.48 (0.82)	1.18 (0.42)	0.78 (0.42)
Raw material III	0.44 (0.14)	97.75 (0.54)	0.71 (0.25)	0.34 (0.15)	0.52 (0.32)	0.21 (0.11)
Raw material IV	1.26 (3.20)	35.85 (8.20)	7.62 (2.91)	42.72 (8.11)	5.02 (2.85)	1.91 (1.17)
Ash content of coal	15.54 (1.36)					
Coal ash		59.77 (3.09)	17.98 (3.01)	10.65 (2.03)	3.18 (0.78)	3.01 (1.37)
Raw material mix (measured)	35.43 (0.34)	13.42 (0.13)	3.17 (0.05)	2.38 (0.05)	41.99 (0.19)	1.90 (0.16)
Clinker (measured)	0.16 (0.04)	21.81 (0.06)	4.90 (0.10)	3.79 (0.10)	64.29 (0.33)	2.82 (0.17)

Note: Standard deviation is displayed in the parentheses.

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