

Feasibility Study of the Use of Concrete Sludge As Alternative Raw Material for Portland Clinker Production

Joris Schoon¹; Klaartje De Buysser²; Isabel Van Driessche³; and Nele De Belie⁴

Abstract: This study aims to examine the use of concrete sludge from water treatment installations of ready-mixed concrete plants as an alternative raw material for portland clinker kilns, and to enumerate possible limitations. By its extensive and representative collection of concrete sludge samples data, it may also be used for other investigations. The chemical variations of this sludge were evaluated by gathering 87 sludge samples over a period of two years. Furthermore, a treatment phase was proposed. Numerical simulations were conducted based on the chemistry of a representative concrete sludge sample, before and after a specific treatment phase, together with classic raw materials of three reference clinker factories to investigate the influence of the sludge on the clinker meal composition. Based on these simulations, experimental clinkers were produced, which were fully analyzed and evaluated for possible mineralogical influences. Based on the findings of this investigation, it was concluded that sludge out of concrete plants could not be classified as a feasible alternative raw material. Nevertheless, this paper has unveiled some attention points for the use of recycled materials or by-products as alternative raw materials for portland clinker production. DOI: [10.1061/\(ASCE\)MT.1943-5533.0001230](https://doi.org/10.1061/(ASCE)MT.1943-5533.0001230). © 2014 American Society of Civil Engineers.

Author keywords: Clinker; Concrete sludge; Water treatment installation; Fines; Sulfur trioxide.

Introduction

The production and transport of ready-mixed concrete necessitate the recuperation of sludge water that is too contaminated with concrete residuals to be discharged into sewerage. This sludge water is generated by washing of the ready-mixed concrete and transport equipment during daily maintenance. Prior to the disposal of sludge water from the ready-mixed concrete plants, a pretreatment in two sedimentation basins is required. The first basin receives the sludge water from the mixer trucks. The water fraction, with some limited sediment fraction, is transferred to a second basin. After sedimentation, water from both basins is recycled and the sludge in the first and the second basins is removed. Globally, the sludge is disposed as landfill, which has an ecological impact and a financial disadvantage in terms of disposal costs in accordance of its Eural-code (101314) (Ökopol GmbH et al. 2008). Therefore, the valorization of this sludge of concrete plant (SCP) is a key concern in the production of ready-mixed concrete and the reason for this feasibility study. Several studies have already examined the recuperation of

different types of sludge and sludge water in fresh concrete, which demonstrated its negative impact on compressive strength and setting time, workability, and water demand (Chatveera et al. 2006; Sales et al. 2011; Chatveera and Lertwattanakul 2009; Lin et al. 2005; Tay 1989; Lin and Lin 2005).

This paper investigates the valorization of the sludge fraction as an alternative raw material (ARM) for portland clinker production, based on an extensive and representative collection of concrete sludge sample data. The study includes a batch of 87 sludge samples from the first basin, which were collected over a period of two years to ensure a realistic evaluation. The collected samples of the dried sludge were analyzed for their chemical composition and variations, and for their physical properties. These parameters are important because they influence the feasibility of using the dry sludge as ARM (Schoon et al. 2012, 2013a, b, unpublished data, 2014). The chemical and mineralogical composition of sludge is primarily influenced by the raw materials used for the production of ready-mixed concrete. The water fraction according to *NBN EN 1008* [Bureau of Normalization (NBN 2002)] can be reused in the ready-mix concrete production process, during which a fraction of small sediments in the water is allowed.

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Note. This manuscript was submitted on April 29, 2014; approved on October 29, 2014; published online on December 24, 2014. Discussion period open until May 24, 2015; separate discussions must be submitted for individual papers. This paper is part of the *Journal of Materials in Civil Engineering*, © ASCE, ISSN 0899-1561/04014272(12)/\$25.00.

Materials and Methods

Classic Raw Materials

The materials used on a daily basis in three modern clinker factories are selected as representative classic raw material (CRM). These factories are CBR Antoining (Ant) and CBR Lixhe (Lxh) in Belgium, and ENCI Maastricht (Maa) in the Netherlands, all belonging to the Heidelberg Benelux group. They can be considered as representative for modern clinker factories worldwide. CBR Antoining uses rich and poor limestone, CBR Lixhe uses tufa and loam, and ENCI Maastricht uses a typical marl and sabulous clay to deliver the required CaO and SiO₂. All three factories use fly ash as the source of Al₂O₃ and an artificially produced Fe₂O₃ source. These CRMs have been described in detail (Schoon et al. 2012)

Table 1. Average Chemical Analysis of the Limestones and SiO₂ Sources of CBR Antoing, CBR Lixhe, and ENCI Maastricht

CRM	Unit	Poor limestone	Rich limestone	Tufa	Loam	Marl	Sabulous clay
CaO	% by weight	42.9	50.1	51.8	5.6	50.8	2.66
SiO ₂	% by weight	15.1	6.4	4.7	68.9	7.1	86.83
Al ₂ O ₃	% by weight	2.2	0.9	0.4	7.4	0.8	3.73
Fe ₂ O ₃	% by weight	0.9	0.4	0.3	3.8	0.4	2.58
K ₂ O	% by weight	0.68	0.21	0.07	1.68	0.13	1.14
Na ₂ O	% by weight	0.25	0.25	0.02	0.71	0.20	0.14
SO ₃	% by weight	0.90	0.57	0.09	0.06	0.21	0.05
MgO	% by weight	1.1	0.9	0.7	0.8	0.8	0.28
Cl	% by weight	—	—	0.011	—	—	0.01
LOI 975°C (O ₂)	% by weight	35.04	40.18	42.03	10.1	40.18	3.43

and used in other investigations (Schoon et al. 2012, 2013a, b). The chemical analyses of the corresponding CRMs are presented in Table 1.

Alternative Raw Material: Recycled Concrete Sludge

A first batch of SCP was collected from 20 ready-mixed concrete plants of Inter Beton (IB), belonging to the Heidelberg Benelux group. This collection was necessary to have an initial idea of the chemical variation of the sludge. From this first batch, five ready-mixed concrete plants were selected according to their geographic position, mutual variation in origin of aggregates (Ag), and the widest possible chemical variation, based on the chemical analyses of the first batch of sludge samples (Fig. 1). IB Genk was selected because the sample from the first batch had the lowest CaO (% by weight) and the highest SiO₂ (% by weight). Additionally, its geographical position in the north-east of Belgium allows IB Genk to partially use river gravel. IB Brussel was chosen because its sample from the first batch had the lowest SiO₂ (% by weight) and it is located in the center of Belgium. Furthermore, IB Brussel regularly works with porphyry material (Schoon et al. 2013b). IB Villers-Le-Bouillet was used as backup for IB Brussel, as the second lowest in SiO₂ (% by weight), because it was communicated that adaptations to the water treatment installation were planned, so

sampling during a certain time was not possible. Another reason was that this plant is located in the east of Belgium and works with dolomitic limestone, which is found locally in the region of Namur (Schoon et al. 2013b). IB Zeebrugge was chosen because it had a similar amount of average SiO₂ (% by weight) in the first batch. This plant uses sea gravel for concrete production, because it is located in the west of Belgium, close to the sea. IB Dendermonde was selected because it has the newest and most modern water treatment installation within the IB group. Just like IB Brussel, IB Dendermonde is located in the center of Belgium and works with a wide variety of aggregates, such as porphyry, limestone, and sea material.

To create a representative batch of sludge samples, a sample was taken each month from the plants of IB Zeebrugge and IB Dendermonde. This made it possible to investigate, if present, the influence of the seasons on the chemical composition of the sludge. The chemical composition could be altered during winter by the increased use of portland cement and during summer by the increased use of slag cement, which are often used in Belgium and the Netherlands. From the plants of IB Brussels, IB Genk, and IB Villers-le-Bouillet, samples were collected every three months. After its water treatment installation was revamped, sampling of IB Brussels was collected each month. This made a total of another

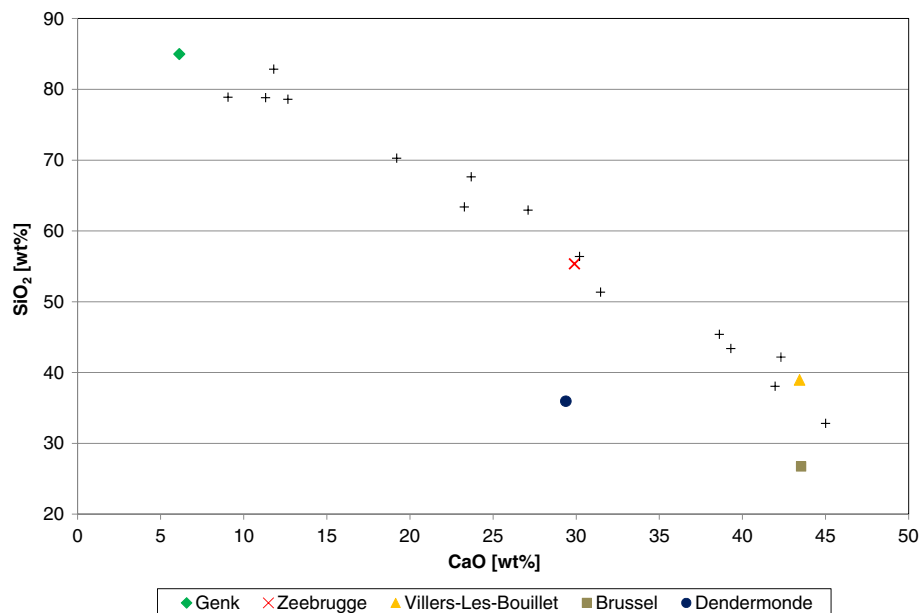


Fig. 1. CaO (% by weight) as function of SiO₂ (% by weight) without LOI, for the first 20 batch sludge samples

67 concrete sludge samples in a second batch. Together with the first batch, 87 sludge samples were collected over a period of two years.

Testing of Raw Materials; Cold Clinker Meals; Clinker and Cement Properties

The procedure to recover the sludge material from the basin was to shovel the concrete sludge with the bucket of a wheel loader ($\pm 2 \text{ m}^3$), deposit the material on a flat floor, and leave it to allow the water to drain away. After 1 h, a sample (5 L) was collected from the middle of the heap and transferred to a bucket. Each month, the different sludge samples of the concrete plants were collected and shipped to the lab, after which they were dried for two days at $120^\circ\text{C} \pm 1$ and crushed to $\pm 4\text{-mm}$ -sized particles in a jaw crusher (Retsch, Haan, Germany) to simulate a realistic crushing action. Five hundred grams were taken from the homogenized sludge sample and further prepared for chemical analysis. The rest of the crushed material was collected to determine the particle size distribution (PSD) by using a sieve tower from $63 \mu\text{m}$ up to 16 mm according to *NBN EN 1097-6 (NBN 2000)*. From the 87 collected concrete sludge samples, 15 samples were selected for determination of PSD. The PSD of the fines fraction collected after the passing the $250\text{-}\mu\text{m}$ sieve was further determined by Sympatec laser diffraction. These 15 samples were chosen by selecting three sludge samples of each of the five concrete plants, assuring satisfactory representation in the sludge samples collected from the five selected concrete plants (Fig. 2). To investigate the effect of a separation phase on the sludge samples already shown as prosperous for production of recycled concrete aggregates as a function of their ARM fitness (Schoon et al., unpublished data, 2014), these 15 samples were recovered after PSD measurement in three fractions: a fines fraction smaller than $250 \mu\text{m}$, a fraction between 250 and $500 \mu\text{m}$, and a fraction larger than $500 \mu\text{m}$. Finally, from these 15 selected samples, the sludge sample marked as “SCP/Avg” (Avg = average) in Fig. 2 was chosen, having SiO_2 (% by weight) and CaO (% by weight) near the average of all 87 collected sludge samples. The samples and separated fractions selected for chemical analysis were prepared by

grinding for 2 min in a vibratory disc mill. X-ray fluorescence (XRF) analysis was performed on a Philips PW2404 and the total carbon (C) and sulfur (S) contents were determined by using a Leco CS230. Thermogravimetric analysis (TGA) and differential thermal analysis (DTA) was performed on a Netzsch STA 449F3. TGA analyses were performed on the separation fractions and the sludge sample itself of five concrete sludge samples individually taken from each of the five concrete plants. They were selected to be most similar to the average chemical composition of their proper concrete plant (Fig. 2) and already present in the collection of the 15 selected concrete sludge samples. SCP/Avg and its fines fraction smaller than $250 \mu\text{m}$ (Fines/Avg) were individually used to make numerical simulations in addition to artificial clinkers for each clinker factory. The different cold clinker meal (CCM) compositions were all prepared as described by Schoon et al. (2012). All raw materials were crushed in a Siebtechnik Disc mill and homogenized in a vessel used for analysis of micro-Deval abrasion resistance, after which they were further ground for 10 min at 300 rpm in a laboratory ball mill. All CCM were, after a granulation phase, sintered in an electric high-temperature static kiln (Carbolite BLF1800) up to $1,450^\circ\text{C}$ at a constant heating rate ($10^\circ\text{C}/\text{min}$). The hot clinker meals (HCMs) were maintained at the scheduled temperature for 1 h after which they were quenched to room temperature to form the final clinker. X-ray diffraction (XRD) analysis, refined by the Rietveld method (*TOPAS* software), was performed on a Bruker D8 ADVANCE. All of these analysis methods have been used and described in more detail in other research papers (Schoon et al. 2012, 2013a, b, unpublished data, 2014).

Theory/Calculation

Chemical and Mineralogical Limitations of Reference Clinkers and Clinker Kiln

Realistically simulated clinker production at lab scale is quite difficult because of the specific construction of a clinker kiln. Therefore, limitations have to be defined that may prevent undesirable

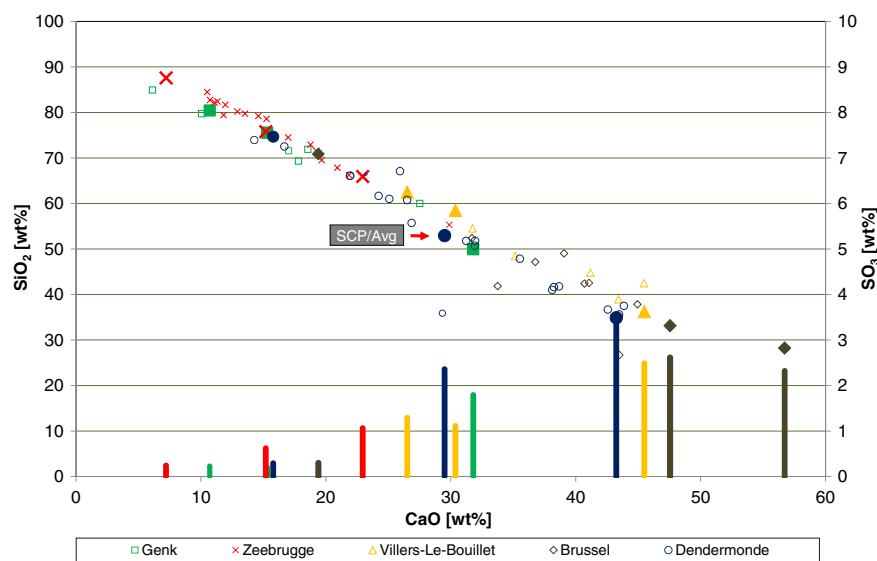


Fig. 2. CaO (% by weight) as function of SiO_2 (% by weight) for the 72 first and second batch sludge samples of the five selected concrete plants and SO_3 (% by weight) without LOI (bars) of the 15 selected sludge samples for PSD (bold)

Table 2. Chemical and Mineralogical Limits of the Final Clinker

Clinker	Unit	Antoing	Lixhe	Maastricht
Cl	% by weight	$x < 0.08$	$x < 0.08$	$x < 0.08$
SO ₃	% by weight	$x < 1.4$	$x < 1.2$	$x < 1.1$
Na ₂ Oeq	% by weight	$x < 1.2$	$x < 1.2$	$x < 1.2$
MgO	% by weight	$x < 4.0$	$x < 4.0$	$x < 4.0$
MgO/Fe ₂ O ₃	—	$x < 1.40$	$x < 1.40$	$x < 1.40$
DOS level	—	$80 < x < 120$	$80 < x < 120$	$80 < x < 120$
If MgO < 2% by weight				
LSF_MgO	—	98.2 ± 0.5	98.19 ± 0.5	98.20 ± 0.5
C ₃ A	% by weight	7.35 ± 0.5	6.65 ± 0.5	7.33 ± 0.5
LiqSimple	% by weight	19.18 ± 0.5	22.73 ± 0.5	22.97 ± 0.5
SR	—	3.2 ± 0.5	2.6 ± 0.5	2.5 ± 0.5

effects on the clinker kiln and insufficient clinker quality based on the cement standards *EN 197-1*, *EN 197-2*, and *EN 196-3* [European Committee for Standardization (CEN) 2000a, b, c]. These limitations have to be taken into account while creating the simulations and corresponding lab clinkers for each clinker kiln/factory. This makes it possible to evaluate the feasibility of applying a different ARM for each clinker kiln/factory.

Therefore, specific chemical limits for SO₃, Cl, alkalis, and MgO for each clinker factory were defined and listed in Table 2. With respect to the mineralogy of the final clinker, limits are also defined for the following three parameters: lime saturation factor (LSF), C₃A, and the liquid phase [simple liquid (LiqSimple)], as defined by Taylor (1997). They are also incorporated in Table 2. The LSF governs the ratios of alite and belite, C₃A describes the expected celite content by means of the well-known Bogue calculation ($2.650 \cdot \text{Al}_2\text{O}_3 - 1.692 \cdot \text{Fe}_2\text{O}_3$), and LiqSimple governs the ratio of Al₂O₃ to Fe₂O₃. By limiting these three parameters for the final clinker, the four metal oxides are sufficiently anchored for each factory. These limits were described in detail by Schoon et al. (2012).

Controlling Parameters for Clinker Feed Calculation

A simulation program based on linear equations was used to calculate CCM for each factory (CCM/Ant, Lxh, Maa) from their corresponding CRMs, in the case of the reference CCM. In the case of the alternative CCM, they were calculated partly from the described ARM. These compositions starting from CRM and ARM were merely adjusted to obtain the mineralogical settings, as already explained, without taking into account the ashes of the fuels that will actually be used to heat up the clinker meal in a real clinker kiln. The alternative CCMs were calculated to maximize the use of SCP/Avg and Fines/Avg. Because of the high SiO₂ (% by weight) in both ARMs, it was expected that they would act as the SiO₂ source in CCM, replacing loam or sabulous clay as classic raw materials (Table 1) in the case of CBR Lixhe and ENCI Maastricht.

Detailed calculations are provided by Schoon et al. (2012). Alkali sulfates decrease the viscosity of the melt, which results in increased alite formation (Taylor 1997). Without sufficient SO₃ (% by weight), the unbound alkali will increase the viscosity of the melt, which conversely results in decreased alite formation (Taylor 1997). To prevent this undesired decrease in alite, a stoichiometric balance is imposed between SO₃ and alkali, expressed as the degree of sulfatization (DOS) value by Hewlett (1988). This value is calculated by Eq. (1), using the chemical analysis of the final clinker

$$\text{DOS} = 77.41 \times \text{SO}_3 / (\text{Na}_2\text{O} + \text{K}_2\text{O} \times 0.658) \quad (1)$$

Currently, DOS levels between 80 and 120% by weight are used in the three clinker factories. Furthermore, it was stated by Taylor (1997) that the burnability of the CCM or the ease by which free lime can be reduced in the kiln to an acceptable value, decreases with increased LSF_MgO [Eq. (2)] or saturation rate (SR) [Eq. (3)]

$$\text{LSF_MgO} = 100 \times (\text{CaO} + 0.75 \times \text{MgO}) / (2.8 \times \text{SiO}_2 + 1.18 \times \text{Al}_2\text{O}_3 + 0.65 \times \text{Fe}_2\text{O}_3) \quad (2)$$

$$\text{SR} = \text{SiO}_2 / (\text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3) \quad (3)$$

By keeping both values constant, the burnability will not be influenced by chemical variations.

Results and Discussion

Evaluation of the Concrete Sludge Samples before Treatment

As shown in Fig. 1, the first batch of concrete sludge samples collected in the beginning of 2011 already showed large chemical variation. The second batch of concrete sludge samples of the five selected concrete plants, collected over a time period (2011–2012) of two years (Fig. 2), which gave a total of 87 samples, had a comparable chemical variation to the first batch. This demonstrates that the 20 samples collected in the beginning of 2011 already provided a representative image of the chemical variation of concrete sludge samples in Belgium.

Second, the chemical variation, although still pronounced, improves visually in Fig. 2 if the sludge is collected for each concrete plant individually. Nevertheless, based on the calculated average, SD, and standard error of the mean (SEM) for both CaO (% by weight) and SiO₂ (% by weight), the improvement of the chemical variation if collected by individually concrete plant is demonstrated to be rather marginal. (Table 3). There was no influence of the seasons observed in the sludge samples of the two concrete plants where samples were collected each month: IB Dendermonde (Fig. 3) and IB Zeebrugge (Fig. 4). Furthermore, a strong relationship was noticed between the chemical composition of the sludge

Table 3. Statistical Interpretation of the Chemical Variations of the Concrete Sludge Samples

Concrete sludge	Unit	Total	IB Brussel	IB Dendermonde	IB Genk	IB Villers-Le-Bouillet	IB Zeebrugge
Amount	—	72	11	23	9	8	21
Average CaO	% by weight	26.74	39.55	30.29	17.21	37.41	16.16
SD CaO	% by weight	12.13	9.57	9.06	8.22	7.42	5.55
SEM CaO	% by weight	1.26	2.89	1.89	2.74	2.63	1.21
Average SiO ₂	% by weight	59.81	42.93	53.03	71.49	48.31	75.44661
SD SiO ₂	% by weight	16.74	12.36	13.32	10.89	9.44	7.98
SEM SiO ₂	% by weight	1.74	3.73	2.78	3.63	3.34	1.74

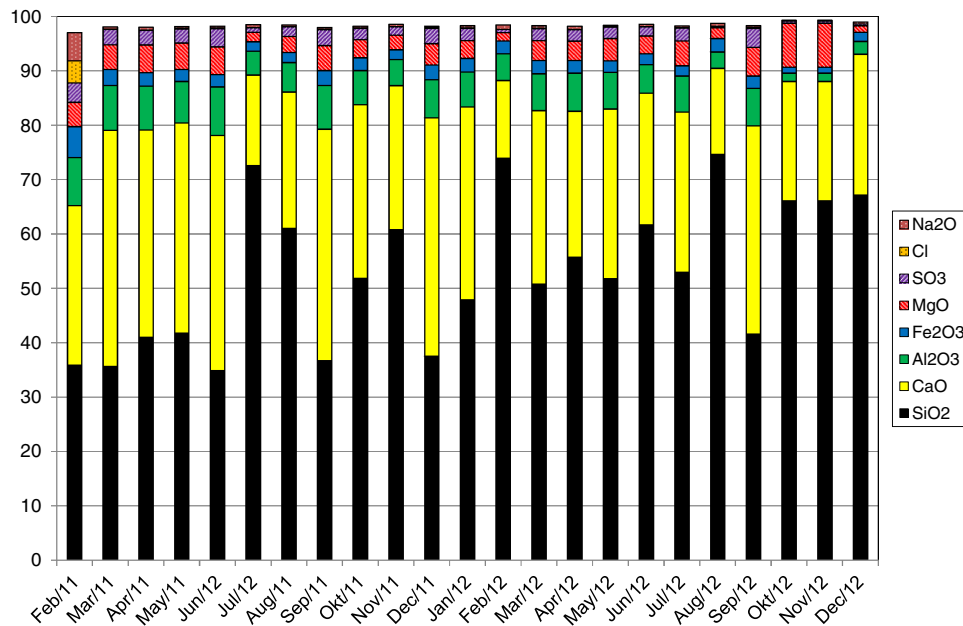


Fig. 3. Chemical composition without LOI of the sludge samples from IB Dendermonde

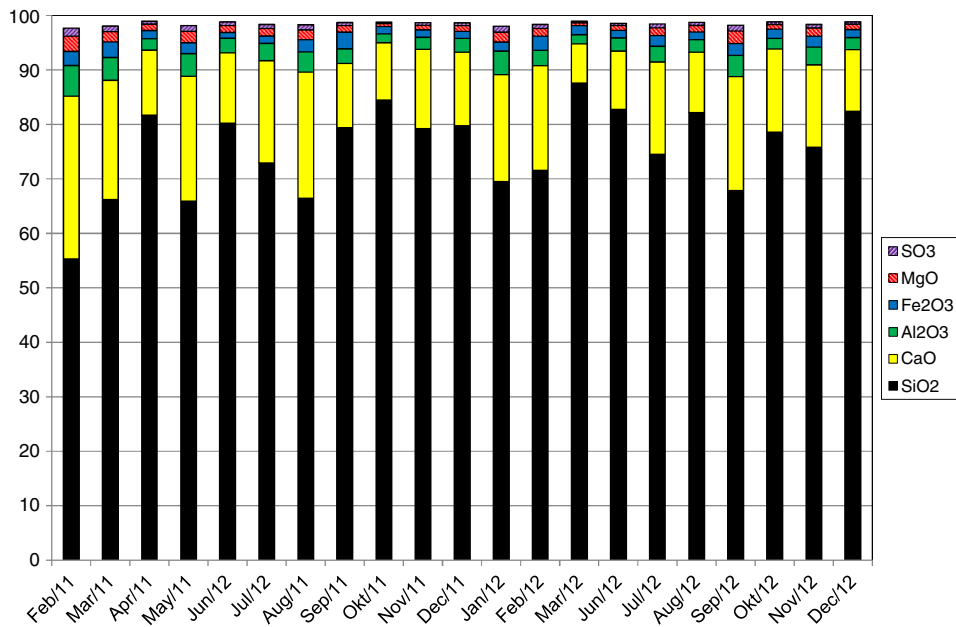


Fig. 4. Chemical composition without LOI of the sludge samples from IB Zeebrugge

and its SO_3 (% by weight). The higher CaO (% by weight) and lower SiO_2 (% by weight) without loss on ignition (LOI) present in the sludge, the higher the SO_3 (% by weight) becomes, as demonstrated in Fig. 2, in which the SO_3 (% by weight) of the 15 selected concrete samples is additionally plotted. This relationship is also demonstrated in Fig. 5, in which CaO (% by weight) of all 87 samples is plotted as a function of their SO_3 (% by weight). Although SO_3 (% by weight) can also be present in the aggregate fraction, especially when sea gravel and sea sand is involved, the largest part of the SO_3 in the sludge is delivered by the cement fraction. As shown in Fig. 5, the SO_3 (% by weight) in concrete sludge can increase to 3.5% by weight, which indicates a high cement content and perhaps an accumulation of the water soluble SO_3 coming from the cement hydrates.

PSD analysis of the 15 selected sludge samples showed also some interesting findings. The upper sieve size (D) of the 15 selected sludge samples is as defined in *NBN EN 1097-6 (2000)* between 4 and 16 mm, and the samples have sand fractions varying from 61 to 100% by weight. Additionally, the fines fraction ($<250 \mu\text{m}$) shows large variation in this pool of 15 samples, from 8 to 58% by weight. When focusing on the fraction smaller than $63 \mu\text{m}$, a variation between 0 and 20% by weight is found in the 15 sludge samples.

TGA analyses clearly show that the five concrete sludge samples have mass losses by dehydration of hydration products between 125 and 650°C and decarbonation peaks between 650 and 900°C when heated (Table 4). The hydration products result from the reacted cement, whereas the carbonated products probably arise

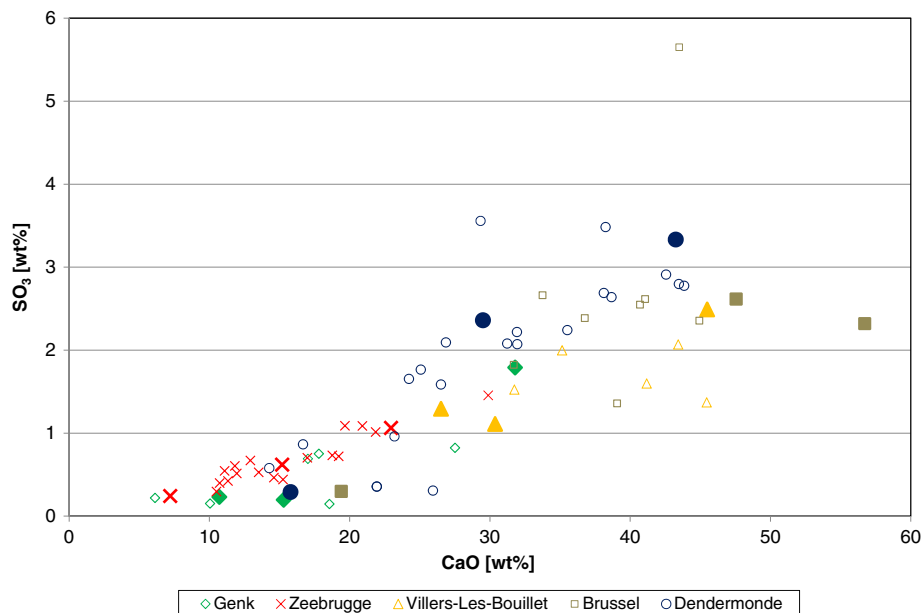


Fig. 5. The relationship between CaO (% by weight) and SO₃ (% by weight) for the 87 concrete sludge samples

from limestone, originating from the cement or the aggregates present in the sludge. The results of this TGA/DTA evaluation are presented in Table 4.

Evaluation of the Concrete Sludge Samples after a Separation Phase

The large chemical variation and high SiO₂ (% by weight) indicate that concrete sludge does not have high potential as ARM for portland clinker production if the concrete sludge cannot be altered. Although it was foreseen that a crushing action combined with a separation phase at 250 μm would separate the hydrated cement from the aggregate fraction, as demonstrated by Schoon et al. (unpublished data, 2014), the chemical composition and chemical

variation did not significantly improve for the recovered fines fraction (<250 μm) (Tables 5 and 6) compared to the total concrete sludge sample (Fig. 2) from which the fines fraction originated. The fines fraction (<250 μm) had similar chemical compositions and variation, and there was a clear relationship between the chemistry (CaO and SiO₂) and SO₃ (% by weight) (Fig. 6), as was also demonstrated for the concrete sludge samples.

TGA analyses of the three separation fractions for the sludge samples similar to the average chemical composition of their concrete plant (Table 4) provide a clear view on the composition of the three separated fractions and the sludge sample itself. As expected, the fraction <250 μm contains cement hydration products visible between 125 and 650°C, as was also observed for recycled concrete

Table 4. Properties of Three Size Fractions of the Five Selected Sludge Samples

Fractions (μm)	Distribution (% by weight)	SiO ₂ (% by weight)	CaO (% by weight)	SiO ₃ (% by weight)	LOI 975°C (O ₂) (% by weight)	125–900°C (% by weight)	125–650°C (% by weight)	650–900°C (% by weight)	Decarb/CaO (% by weight)
Brussel	100	22.14	44.49	1.82	21.56	-22.12	-9.15	-12.97	-0.29
>500	66	15.89	46.17	1.27	28.93	-24.41	-7.46	-16.95	-0.37
250 < x < 500	9	37.80	37.45	1.24	16.55	-16.30	-10.91	-5.39	-0.14
<250	25	22.41	41.34	2.40	21.91	-18.15	-12.96	-5.19	-0.13
Dendermonde	100	45.53	25.37	2.03	14.00	-14.34	-10.83	-3.52	-0.14
>500	43	46.74	24.53	2.30	14.22	-11.91	-8.89	-3.02	-0.12
250 < x < 500	6	73.09	11.64	1.01	7.34	-6.25	-4.55	-1.70	-0.15
<250	51	40.51	26.55	2.21	17.43	-17.35	-13.20	-4.15	-0.16
Genk	100	67.04	13.57	0.17	11.27	-11.21	-2.69	-8.52	-0.63
>500	80	65.73	14.55	0.13	11.82	-11.71	-1.82	-9.89	-0.68
250 < x < 500	12	80.34	6.27	0.21	4.99	-3.54	-1.35	-2.19	-0.35
<250	8	53.08	17.90	1.17	13.51	-17.71	-13.37	-4.34	-0.24
Villers-Le-Bouillet	100	48.04	24.94	0.91	17.87	-17.79	-8.72	-9.07	-0.36
>500	40	37.39	30.52	0.92	22.94	-18.37	-5.72	-12.65	-0.41
250 < x < 500	16	82.77	6.59	0.21	4.88	-4.83	-1.43	-3.39	-0.51
<250	44	30.90	31.53	1.77	23.02	-21.98	-14.10	-7.89	-0.25
Zeebrugge	100	83.12	6.84	0.23	5.11	-5.67	-2.85	-6.68	-0.98
>500	49	71.27	13.27	0.37	9.86	-6.64	-3.94	-10.57	-0.80
250 < x < 500	31	88.20	4.53	0.10	3.37	-2.28	-0.10	-2.18	-0.48
<250	20	74.25	10.62	0.53	6.49	-8.57	-4.46	-4.11	-0.39

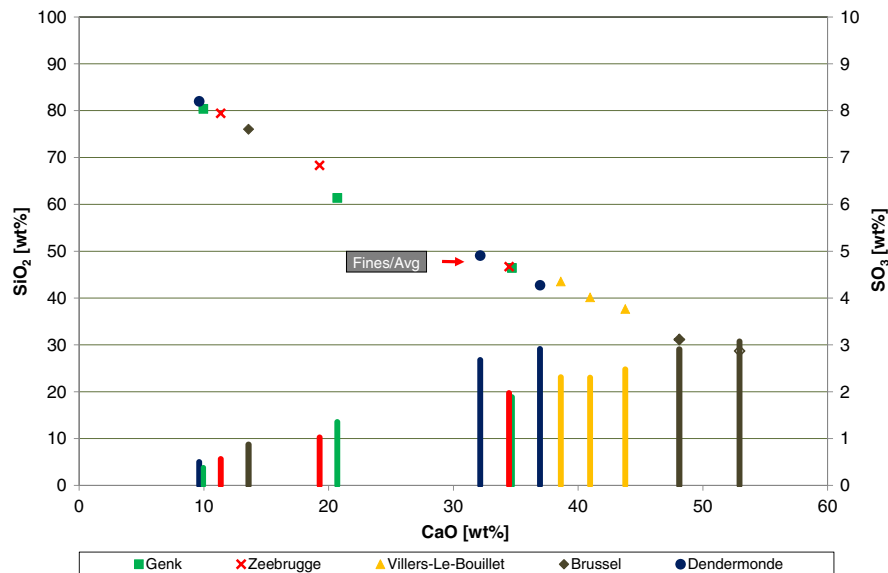
Note: 125–650°C = hydration products; 650–900°C = decarbonation; Decarb/CaO = 650–900°C/CaO; boldface indicates the complete sludge samples.

Table 5. Average Chemical Compositions of the Three Selected Sludge Samples from Each Concrete Plant

ARM	Unit	Brussel	Dendermonde	Genk	Villers-Le-Bouillet	Zeebrugge
		Average	Average	Average	Average	Average
CaO	% by weight	33.08	24.53	17.01	27.76	13.54
SiO ₂	% by weight	36.78	46.60	61.67	43.14	69.75
Al ₂ O ₃	% by weight	4.54	5.14	3.98	4.26	2.71
Fe ₂ O ₃	% by weight	1.80	1.89	2.62	1.55	1.72
K ₂ O	% by weight	0.53	0.52	0.53	0.50	0.47
Na ₂ O	% by weight	0.37	0.34	0.22	0.14	0.37
SO ₃	% by weight	1.39	1.64	0.65	1.32	0.57
MgO	% by weight	2.44	3.23	2.06	2.40	1.23
TiO ₂	% by weight	0.32	0.32	0.27	0.32	0.15
P ₂ O ₅	% by weight	0.15	0.15	0.14	0.12	0.13
Cl	% by weight	0.04	0.06	0.02	0.05	0.07
LOI 975°C (O ₂)	% by weight	18.25	15.24	10.59	18.15	9.10

Table 6. Average Chemical Compositions of the Three Selected Sludge Fines from Each Concrete Plant

ARM	Unit	Brussel	Dendermonde	Genk	Villers-Le-Bouillet	Zeebrugge
		Average	Average	Average	Average	Average
CaO	% by weight	30.02	21.85	18.74	31.56	18.86
SiO ₂	% by weight	38.23	50.78	55.85	31.08	58.10
Al ₂ O ₃	% by weight	4.91	5.12	5.13	5.48	4.70
Fe ₂ O ₃	% by weight	2.03	1.61	2.71	1.79	1.84
K ₂ O	% by weight	0.54	0.66	0.69	0.47	0.73
Na ₂ O	% by weight	0.28	0.31	0.27	0.16	0.52
SO ₃	% by weight	1.80	1.68	1.03	1.81	1.03
MgO	% by weight	2.67	3.10	2.79	3.46	2.20
TiO ₂	% by weight	0.36	0.32	0.37	0.45	0.26
P ₂ O ₅	% by weight	0.20	0.16	0.17	0.15	0.17
Cl	% by weight	0.04	0.07	0.02	0.07	0.13
LOI 975°C (O ₂)	% by weight	18.60	14.08	11.96	23.19	11.24

**Fig. 6.** Fines (<250 μm) of the 15 sludge samples from the five selected concrete plants

(Schoon et al., unpublished data, 2014). Interestingly, the >500 μm fraction in each of the 15 analyzed sludge samples contained comparable cement hydration products and have comparable CaO (% by weight) and SiO₂ (% by weight). The fraction between 250 and 500 μm consisted of a low amount of cement hydrates.

TGA analysis between 650 and 900°C demonstrates less decarbonation of CaCO₃ (% by weight) in the <250 μm fraction than in the corresponding sludge sample itself. By presenting the decarbonation between 650 and 900°C as a function of CaO (% by weight) (Decarb/CaO), it is clear that the <250 μm fraction consists of less

Table 7. Chemical Compositions of the Three Separation Fractions of SCP/Avg

Fractions (μm)	Unit	Fraction	Fraction	Fines/Avg	SBC/Avg
		>500	$250 < x < 500$	<250	Total
% by weight	—	43	6	51	100
CaO	% by weight	24.53	11.64	26.55	25.37
SiO ₂	% by weight	46.74	73.09	40.51	45.53
Al ₂ O ₃	% by weight	5.55	3.04	5.81	5.66
Fe ₂ O ₃	% by weight	1.51	0.87	1.60	1.65
K ₂ O	% by weight	0.53	0.49	0.59	0.54
Na ₂ O	% by weight	0.30	0.21	0.24	0.26
SO ₃	% by weight	1.97	1.01	2.21	2.03
MgO	% by weight	3.76	1.87	4.08	3.93
TiO ₂	% by weight	0.37	0.19	0.42	0.41
P ₂ O ₅	% by weight	0.18	0.09	0.18	0.17
Cl	% by weight	0.05	0.03	0.07	0.06
LOI 975°C (O ₂)	% by weight	14.22	7.34	17.43	14.00

Table 8. Compositions of the Different Clinker Meals Created to be Fed to the Static Kiln

CCM/Ant	CRM (Ant) + ARM	Quantity (% by weight)	CCM/Lxh	CRM (Lxh) + ARM	Quantity (% by weight)	CCM/Maa	CRM (Maa) + ARM	Quantity (% by weight)
CCM/Ant/Ref	Poor limestone	55.25	CCM/Lxh/Ref	Tufa	79.44	CCM/Maa/Ref	Marl	84.38
	Rich limestone	37.50		Loam	6.63		Sabulous clay	2.90
	Fly ash	6.38		Fly ash	12.34		Fly ash	11.02
	Iron carrier	0.87		Iron carrier	1.59		Iron carrier	1.70
	SCP/Avg	0.00		SCP/Avg	0.00		SCP/Avg	0.00
	Fines/Avg	0.00		Fines/Avg	0.00		Fines/Avg	0.00
	Sum	100.00		Sum	100.00		Sum	100.00
CCM/Ant/SCP	Poor limestone	32.76	CCM/Lxh/SCP	Tufa	75.20	CCM/Maa/SCP	Marl	77.14
	Rich limestone	54.71		Loam	2.01		Sabulous clay	1.62
	Fly ash	6.74		Fly ash	11.36		Fly ash	8.61
	Iron carrier	0.91		Iron carrier	1.80		Iron carrier	1.68
	SCP/Avg	4.87		SCP/Avg	9.64		SCP/Avg	10.94
	Fines/Avg	0.00		Fines/Avg	0.00		Fines/Avg	0.00
	Sum	100.00		Sum	100.00		Sum	100.00
CCM/Ant/Fines	Poor limestone	5.91	CCM/Lxh/Fines	Tufa	75.31	CCM/Maa/Fines	Marl	76.32
	Rich limestone	70.46		Loam	0.00		Sabulous clay	0.00
	Fly ash	6.36		Fly ash	12.78		Fly ash	10.31
	Iron carrier	0.97		Iron carrier	1.54		Iron carrier	1.56
	SCP/Avg	0.00		SCP/Avg	0.00		SCP/Avg	0.00
	Fines/Avg	16.30		Fines/Avg	10.37		Fines/Avg	11.80
	Sum	100.00		Sum	100.00		Sum	100.00

CaCO₃ than the sludge sample itself. The <250 μm fraction will decrease decarbonation in the CCM more than its corresponding sludge sample when equally dosed to an alternative CCM. It is the present cement stone derivatives, and not the total CaO (% by weight), although related, that will determine the potential of concrete sludge to reduce the decarbonation in the CCM. This will be further demonstrated in the next sections. The fraction between 250 and 500 μm consists of an excess of SiO₂ (% by weight) demonstrating the presence of quartz particles and much lower CaO (% by weight) and SO₃ (% by weight), which indicates, together with the lower mass losses between 125 and 650°C, that smaller cement fractions are present than in the >500 and <250 μm fractions. These quartz particles have their origin from the fraction of aggregates of the concrete sludge. The slightly higher SiO₂ (% by weight) in the >500 μm fraction originates from the quartz particles that were also present in the fraction between 250 and 500 μm . To demonstrate this trend in more detail, a full chemical analysis and a TGA analysis of the three fractions of the sludge sample SCP/Avg is presented in Table 7 and Fig. 7.

The findings in this section indicates that a crushing action on 4 mm and separation on 250 μm particles is not sufficient to recover all hydrated cement in the <250 μm fraction, as was expected, and which was demonstrated successfully to recycle fines into ARM generated from recycled concrete aggregates, as demonstrated by Schoon et al. (unpublished data, 2014). One explanation could be that the composition of concrete sludge is not comparable with that of recycled concrete (Schoon et al., unpublished data, 2014). It is composed of the fine particles of the different raw materials (cement, sand, raw aggregates, and fly ash) used to produce concrete. This explains why hydrated cement is much more present in the sludge than in the recycled concrete (Schoon et al., unpublished data, 2014). The increased presence of the hydrated cement and the lack of an aggregate structure negatively influence the crushing action. The cement present in the concrete sludge will have reacted with an excess of water in the concrete sludge basin before it is collected, making the cement stone in the sludge sample very weak after hardening. In this way, the cement stone crumbles instead of breaking apart, leaving many cement stone particles in the fraction

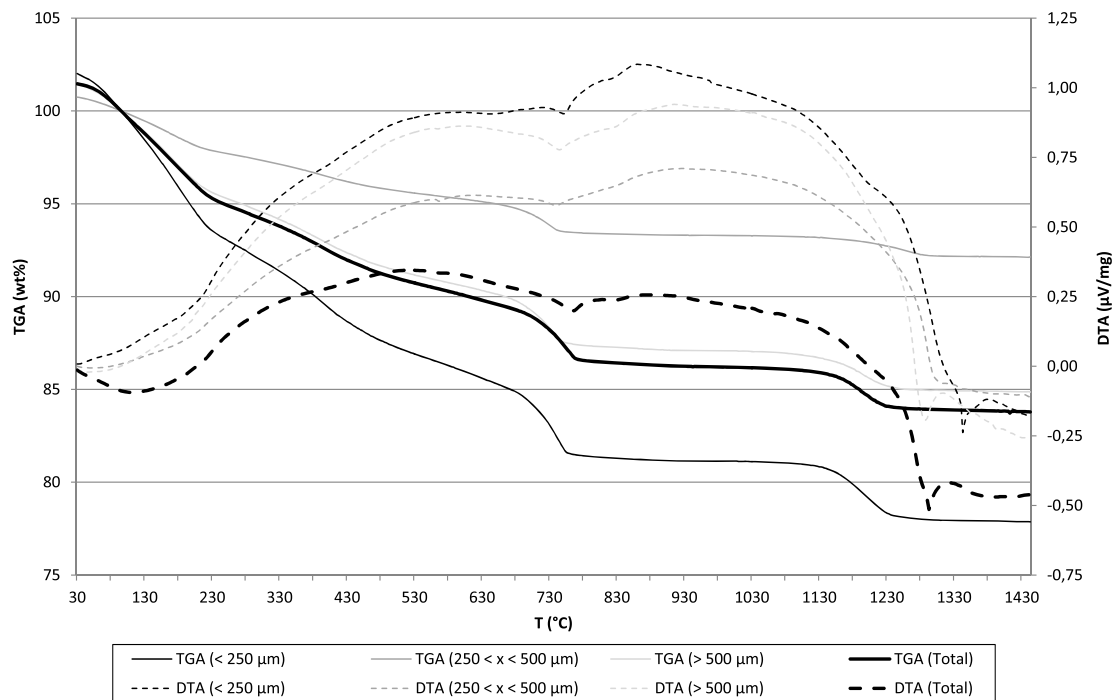


Fig. 7. TGA/DTA analysis of SCP/Avg and its three fractions after a separation phase

larger than 500 μm . Therefore, more intensive crushing action is necessary to separate the hydrated cement stone fraction from the aggregates fraction, although questions may be raised about the economic feasibility and necessity of this intensified crushing action. One reason to strongly consider this intensified crushing action could be the grindability of the coarse quartz particles (Schoon et al., unpublished data, 2014; Kakali and Tsvivilis 1993; Tsamatsoulis 1979; Christensen 1979; Ghosh 1991) present in fractions $250 < x < 500 \mu\text{m}$ and $>500 \mu\text{m}$ and their influence on the burnability of the clinker. This was further investigated and discussed in the next sections.

Clinker Feed Calculations and Preparations

Although the chemical compositions of SCP/Avg and Fines/Avg are comparable, the possible influence on the burnability by the presence of the coarse quartz particles in SCP/Avg could be investigated when sintering alternative CCM with both materials. Therefore, they were both used as ARMs for the calculation and preparation of clinker feed.

As demonstrated in Table 8, Fines/Avg completely replaced the classic SiO_2 source in CCM/Lxh/Fines and CCM/Maa/Fines. The maximization of SCP/Avg was limited by the required CaO (% by weight) defined by LSF and LiqSimple of both kilns; therefore, it only partly replaced the complete SiO_2 source. Because the SiO_2 (% by weight) of the ARM is lower than those present in the classic SiO_2 sources (Table 1), higher volumes of the ARMs have to be dosed to attain the required SiO_2 (% by weight). To compensate for these higher volumes, limestone dosages are decreased. Because less limestone (Table 1) is dosed, CaO (% by weight) in the CCM must be compensated by the CaO (% by weight) present in the ARMs. In the case of CBR Antoining, the influence on CaO (% by weight) acted as a limitation in both alternative CCMs (ARM/Ant/SCP and ARM/Ant/Fines). Maximization of the two different ARM materials is different than for the two previous factories because CBR Antoining does not use a real SiO_2 source. The necessary SiO_2 is delivered by their two limestone sources, particularly poor limestone (Table 1). Fines/Avg, owing to their

higher CaO (% by weight) and lower SiO_2 (% by weight), can be dosed in higher quantities than SCP/Avg.

Evaluation of the Artificially Produced Portland Clinkers

The alternative CCMs show small but quantifiable decreases in decarbonation mass loss (Anorg CO_2) compared to their reference (Table 9), which are more pronounced when using Fines/Avg than the corresponding SCP/Avg. The reason for this difference was already explained in the previous sections. An estimation was made of the measurement of decarbonation mass loss to demonstrate the CaCO_3 (% by weight) present in the reference and alternative CCM, and to quantify the required decarbonation energy (Decarb E CaCO_3 Der) from the known reaction enthalpy of 1,782 kJ/kg for CaCO_3 stated by Taylor (1997). This decarbonation energy can also be measured by DTA analysis, as presented in Table 9. Both methods demonstrate a decrease in decarbonation energy in the alternative CCMs compared to their reference.

The XRD analysis with Rietveld refinement of the final clinkers, presented in Tables 10 and 11, shows mineralogical weight percentages that are on the same order of magnitude, but nevertheless

Table 9. TGA/DTA Evaluation of Reference and Alternative CCMs of CBR Antoining, CBR Lixhe, and ENCI Maastricht

CCM	Anorg CO_2 TGA Meas (% by weight)	CaCO_3 TGA Der (% by weight)	Decarb E	
			Decarb E $\mu\text{Vs/mg}$ Mat DTA	CaCO_3 Der J/g Mat TGA
Mat				
CCM/Ant/Ref	34.0	77.3	213.4	1,378
CCM/Ant/SCP	33.0	75.0	172.2	1,337
CCM/Ant/Fines	30.4	69.1	140.3	1,232
CCM/Lxh/Ref	32.5	73.9	189.3	1,318
CCM/Lxh/SCP	31.7	72.1	176.4	1,285
CCM/Lxh/Fines	31.6	71.8	161.9	1,281
CCM/Maa/Ref	34.2	77.7	219.3	1,385
CCM/Maa/SCP	32.6	74.1	181.6	1,321
CCM/Maa/Fines	31.8	72.3	171.1	1,289

Table 10. Mineralogical Analysis by XRD of the Final Reference Clinkers Produced in a Static Kiln

Clinker	Unit	Cl/Ant/Ref	Cl/Lxh/Ref	Cl/Maa/Ref
Alite (C ₃ S)	% by weight	64.52	65.04	71.33
Belite (C ₂ S)	% by weight	19.73	14.93	8.56
Aluminate (C ₃ A)	% by weight	1.79	3.68	4.64
Ferrite (C ₄ AF)	% by weight	12.86	15.87	14.89
Free Lime (CaO)	% by weight	0.23	0.23	0.35
Periclase (MgO)	% by weight	0.39	0.18	0.23
Arcanite (K ₂ SO ₄)	% by weight	0.32	0.07	—
Aphthitalite	% by weight	—	—	—

different, than those calculated by the Bogue equations, based on the chemical analysis of the final clinkers presented in Tables 12 and 13. The first reason for this deviation is that the DOS factors, although calculated between 80 and 120, did not attain this goal for the final clinkers of CBR Lixhe and ENCI Maastricht. The reason of this unbalance between alkali and SO₃ (% by weight) is attributable to the different volatility of the SO₃ and alkali in a static lab furnace compared to a real clinker kiln, which was explained by Schoon et al. (2012). For both the alternative clinkers of CBR Lixhe and ENCI Maastricht, DOS factors were too low and resulted in lower alite (% by weight) measured by XRD than that calculated by Bogue (Schoon et al. 2012, 2013a, b, unpublished data, 2014). The lower the DOS factor, the greater the difference between the alite (% by weight) measured by XRD and the alite (% by weight) calculated by Bogue, which was extensively explained by Schoon et al. (2012). The alternative clinker (Cl/Ant/SCP Fines) partly produced with the fines fraction (Fines/Avg) had a DOS factor slightly higher (136.35) than the DOS limits and a difference of approximately 4% by weight. Because the high maximization of the fines fraction (Fines/Avg) was approximately 16% by weight, a higher SO₃ (% by weight) than the original sludge sample (SCP/Avg) was introduced into the alternative CCM. Second, by the increased presence of MgO (2.41% by weight) (Table 13) introduced by Fines/Avg (Table 7), increased periclase (% by weight) was measured compared to Cl/Ant/reference (Ref) (Table 10) and Cl/Ant/SCP (Table 11), which partly replaces alite Schoon et al. (2013b). Together with the unbalanced DOS factor, this may explain the deviation measured in Cl/Ant/Fines. Furthermore, the deviation for the alternative clinkers increases with increased dosage (% by weight) of SCP/Avg. As presented in Table 11, higher levels (% by weight) of SCP/Avg also increase the presence of free lime (% by weight), indicating a reduced burnability of the clinker, which was described by Schoon et al. (2013a). Therefore, it can be concluded that the presence of quartz in the >250 μm fraction, as already mentioned, has a negative

Table 12. Chemical Analysis and Bogue Calculations of the Final Reference Clinkers Produced in a Static Kiln

Clinker	Unit	Cl/Ant/Ref	Cl/Lxh/Ref	Cl/Maa/Ref
CaO	% by weight	65.90	66.28	66.18
SiO ₂	% by weight	22.27	21.93	21.39
Al ₂ O ₃	% by weight	4.14	4.40	4.54
Fe ₂ O ₃	% by weight	3.02	4.21	3.98
K ₂ O	% by weight	0.59	0.21	0.33
Na ₂ O	% by weight	0.17	0.20	0.21
SO ₃	% by weight	0.89	0.12	0.36
MgO	% by weight	1.73	1.28	1.52
TiO ₂	% by weight	0.25	0.30	0.30
P ₂ O ₅	% by weight	0.21	0.24	0.17
Cl	% by weight	—	—	—
LOI 975°C (O ₂)	% by weight	0.48	0.39	0.48
DOS factor	—	123.42	27.47	65.24
Alite (C ₃ S)	% by weight	66.84	67.52	70.61
Belite (C ₂ S)	% by weight	13.44	11.95	8.07
Aluminate (C ₃ A)	% by weight	5.86	4.54	5.30
Ferrite (C ₄ AF)	% by weight	9.19	12.81	12.11

effect on the burnability of the clinker. It can be concluded that if sludge of a concrete plant is used as ARM for portland clinker production, low dosages have to be maintained owing to the presence of quartz. A separation phase of 250 μm may counter this problem and allow an increased ARM dosage. Nevertheless, the presence of minor constituents as alkali, SO₃, and MgO has to be closely monitored to minimize their negative influences on the portland clinker process.

Further Investigations to Improve the Feasibility of Concrete Sludge As ARM for Portland Clinker

As was demonstrated in Figs. 1, 2, and 5, there is a clear relationship between CaO (% by weight) and SO₃ (% by weight). This means that if concrete sludge materials can be recovered with the same SO₃ (% by weight), they will have comparable chemical compositions. An investigation will be started to evaluate whether the SO₃ (% by weight) of concrete sludge can be feasibly measured and if the SO₃ (% by weight) of concrete sludge follows a trend in time by which it increases, decreases, or is randomly dispersed. When a clear trend is noticeable, the recovery of the concrete sludge may be individually managed for each concrete plant based on the SO₃ (% by weight), by which a decrease in the chemical variation of the concrete sludge could be possible. Further investigation should demonstrate whether this decrease in chemical variation would be sufficient, together with the findings in the previous sections, to make the sludge acceptable as a valid ARM for portland clinker production.

Table 11. Mineralogical Analysis by XRD of the Final Alternative Clinkers Produced in a Static Kiln

Clinker	Unit	Cl/Ant/SCP	Cl/Ant/Fines	Cl/Lxh/SCP	Cl/Lxh/SCP Fines	Cl/Maa/SCP	Cl/Maa/Fines
Alite (C ₃ S)	% by weight	65.37	56.88	61.54	51.99	61.86	60.86
Belite (C ₂ S)	% by weight	20.32	27.56	20.69	29.06	19.53	19.36
Aluminate (C ₃ A)	% by weight	1.84	3.78	2.30	2.03	2.61	3.15
Ferrite (C ₄ AF)	% by weight	12.00	10.42	14.59	16.03	14.18	15.66
Free lime (CaO)	% by weight	0.06	0.04	0.25	0.09	0.85	0.09
Periclase (MgO)	% by weight	0.36	1.08	0.48	0.40	0.50	0.50
Arcanite (K ₂ SO ₄)	% by weight	—	—	—	0.02	—	0.05
Aphthitalite	% by weight	—	0.09	—	0.13	0.05	0.08
Anhydrite	% by weight	—	—	—	—	0.12	0.02
Portlandite	% by weight	0.06	0.12	0.03	0.24	0.31	0.22

Table 13. Chemical Analysis and Bogue Calculations of the Final Alternative Clinkers Produced in a Static Kiln

Clinker	Unit	Cl/Ant/SCP	Cl/Ant/Fines	Cl/Lxh/SCP	Cl/Lxh/Fines	Cl/Maa/SCP	Cl/Maa/Fines
CaO	% by weight	66.30	65.33	66.08	65.58	66.64	65.98
SiO ₂	% by weight	22.60	22.58	22.29	22.82	22.12	22.20
Al ₂ O ₃	% by weight	4.04	4.40	4.13	4.45	4.11	4.50
Fe ₂ O ₃	% by weight	2.92	2.86	3.87	3.70	3.53	3.66
K ₂ O	% by weight	0.36	0.30	0.21	0.12	0.22	0.23
Na ₂ O	% by weight	0.17	0.20	0.18	0.15	0.19	0.21
SO ₃	% by weight	0.57	0.70	0.24	0.17	0.21	0.20
MgO	% by weight	1.79	2.41	1.65	1.71	1.77	1.81
TiO ₂	% by weight	0.26	0.29	0.29	0.29	0.26	0.29
P ₂ O ₅	% by weight	0.20	0.21	0.23	0.23	0.19	0.17
Cl	% by weight	N/A	0.03	N/A	N/A	N/A	N/A
LOI 975°C (O ₂)	% by weight	0.34	0.33	0.40	0.38	0.35	0.31
DOS factor	—	108.44	136.35	58.39	57.48	48.56	42.85
Alite (C ₃ S)	% by weight	66.77	60.65	66.27	58.30	70.47	64.36
Belite (C ₂ S)	% by weight	14.43	18.00	13.92	21.45	10.27	15.10
Aluminate (C ₃ A)	% by weight	5.77	6.82	4.40	5.53	4.92	5.73
Ferrite (C ₄ AF)	% by weight	8.89	8.70	11.78	11.26	10.74	11.14

Conclusions

A large chemical variation between different concrete sludge samples was noted, which only marginally decreased when the concrete sludge samples were collected individually for each concrete plant. No clear influence of the seasons was observed in the chemical composition of the sludge samples. The chemical variation of concrete sludge is too large to be used as a valid ARM in portland clinker production. It was also shown that the sludge from the water treatment system of concrete plants specifically collects the finer fractions of the various raw materials of the concrete, because of which the cement content in the sludge samples is much higher than in normal recycled concrete. Because of this difference, promising techniques for creating an ARM out of recycled concrete were not successful for concrete sludge. A separated fines fraction of 250 μm does not have improved chemical composition or variation, but has improved burnability within the alternative portland clinker. Furthermore, the higher concentration of cement stone derivatives present in the fines fraction improves the potential to reduce the inorganic CO₂ emission.

Nevertheless, a fraction larger than 250 μm shows no improved suitability for concrete applications because of the presence of high levels of cement derivatives, as was the case for recycled concrete crushed and separated with the same technique.

The presence and variation in concentration of minor constituents, such as alkali, SO₃, and MgO, act as a second limit on the maximization of both sludge samples and their fractions of specific fines.

The possible energy gain from the lowered decarbonation energy attributable to a decreased limestone dosage is negligible because of the energy required for the liberating the chemically bound water. The only positive effect on the ecological impact of portland clinker production is that a primary natural material may be replaced by a secondary recycled material.

After this study, it can be concluded that sludge out of concrete plants cannot be classified as a feasible alternative raw material for portland clinker production owing to the cited restrictions.

Acknowledgments

The authors wish to thank the Central lab of Sagrex Quenast and the cement research lab of ENCI Maastricht for their support, Dr. Els Bruneel for her aid during TGA analysis, Jo Lejeune for

his help during the XRD tests and the staff of the concerned concrete plants for their efforts during the collection of the 87 concrete sludge samples.

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