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## Impact of quicklime reactivity and origin on Autoclaved Aerated Concrete production

**ABSTRACT:** Quicklime is a key component of Autoclaved Aerated Concrete (AAC), with typical quicklime contents of order 15 wt.% in Europe. It is the mostly used source of calcium oxide that eventually ends up as tobermorite in the final product.

In this paper, we describe the research performed at Lhoist in order to assess the influence of quicklime on AAC. Several types of quicklimes were used, differing by their origin and reactivity. They were incorporated together with other fixed ingredients (cement, quartz,...) in several AAC recipes with final densities ranging from 350 to 550 kg/m<sup>3</sup>, corresponding to the European P2 – 0.35, P2 – 0.4 and P4 – 0.55 classes (EN 771-4).

The study consisted first in following the green cake expansion of the different recipes. Then, the green cakes were autoclaved for 10 hours at 11 bars of steam pressure. The obtained AAC blocks were then tested for compressive strength and density.

The study showed that quicklime reactivity had low effect on green cake penetration value. However, quicklime reactivity had a major effect on green cake expansion. Too reactive a quicklime generated a poor pore structure as detected by a fallback effect and a below-specification compressive strength of the low density AAC. Then, quicklime slaking curve was not sufficient to anticipate the green cake expansion for a given recipe. Quicklimes with similar reactivity but different origin could have very different behaviour in the AAC. Finally, the behaviour of the too-reactive quicklimes would probably have been easily corrected by decreasing the water temperature.

### 1. Introduction

Quicklime is a key component of Autoclaved Aerated Concrete (AAC), with typical quicklime contents of order 15 wt.% in Europe. It is the mostly used source of calcium oxide that eventually is transformed into tobermorite as the final product [1].

The role of quicklime is threefold: First, it is the main calcium oxide source that eventually combines with silicates and water to form a mixture of Calcium Silicate Hydrates (C-S-H) phases, among which is tobermorite, the mineral responsible for the AAC strength. Second, quicklime reacts with aluminium allowing for the hydrogen release that generates green cake expansion. Third, quicklime hydrates and forms portlandite through an exothermic reaction whose energy input speeds up the strengthening processes in the green cake initial setting.

In this paper, we describe the research performed at Lhoist in order to assess the influence of quicklime on AAC. For the purpose several types of quicklimes were used, differing by their origin and reactivity. Three European limestone origins were used. For one of the limestone origin, different quicklime reactivities were obtained by changing the calcination conditions at both laboratory and industrial scales.

These various quicklimes were incorporated together with other fixed ingredients (cement, quartz sand,...) in several AAC recipes with final densities ranging from 350 to 550 kg/m<sup>3</sup>, corresponding to the European P2 – 0.35, P2 – 0.4 and P4 – 0.55 classes (EN 771-4).

The study embraced first in following green cake expansion of the different recipes. The monitoring was based on temperature and height. Green cake strength was recorded at the end of the expansion phase. Then, the green cakes were autoclaved for 10 hours at 11 bars of steam pressure. The compressive strength and density of obtained AAC blocks were then tested.

### 2. Experimental

#### 2.1. Materials

##### 2.1.1. Raw materials

The following raw materials for AAC manufacturing were used (Table 1):

- Quartz sand Millisil W 10 from Quarzwerke in Frechen (Germany),
- Portland cement CEM I 42.5 R Wittekind from the PLZ-plant in Erwitte (Germany),
- Anhydrite from Südharzer Gipswerke GmbH (Germany),
- Aluminium paste Aquapor 9004 from Schlenck (Germany).

Several quicklimes were studied (Table 2). One first set of materials was based on quicklimes that were laboratory-calcined at varying temperatures from the same limestone A. This allowed obtaining varying reactivities with a constant chemical composition (quicklimes LA1, LA2 and LA3). Unfortunately, only limited amounts of LA3 were available and only one AAC recipe could be

manufactured of it. Another set of materials was obtained using two quicklimes with different reactivities also made from limestone A but this time at industrial scale (quicklimes IA1 and IA2). Finally, two other industrial quicklimes made in other Lhoist plants of the local limestone were studied (IB and IC). Details on limestone calcination, quicklime manufacturing process and properties can be found in the literature [2].

Table 1

RAW MATERIALS COMPOSITION AS MEASURED BY X-RAY FLUORESCENCE (EXPRESSED IN WT.% OF EQUIVALENT OXIDE).

	Supplier	CaO	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	SO <sub>3</sub>
Cement (CEM I 42.5 R)	Wittekind	65.3	22.3	3.6	3.2
Quartz Sand	Quarzwerte Frechen	--	99.0	--	--

Table 2

THE VARIOUS QUICKLIMES USED IN THE STUDY. CAO AS MEASURED BY X-RAY FLUORESCENCE (EXPRESSED IN WT.% OF EQUIVALENT OXIDE). REACTIVITY (T60) MEASURED BY EN459-2.

	Calcination	Limestone Origin	CaO	t <sub>60</sub>
			wt.%	min
LA1	Laboratory	A	95.5	4.1
LA2	Laboratory	A	95.6	5.2
LA3	Laboratory	A	96.2	7.6
IA1	Industrial	A	93.5	1.9
IA2	Industrial	A	94.0	3.1
IB	Industrial	B	95.8	8.2
IC	Industrial	C	93.8	5.2

### 2.1.2. AAC recipes

Three AAC recipes were used throughout the study. Prior to any AAC manufacturing, return slurry was prepared with the composition described in Table 3. It was maintained under mild agitation (300 rpm) for at least 4 hours before being mixed with the other ingredients.

Green cakes were obtained by blending the return slurry (Table 3) and additional raw materials according to the three recipes described in Table 4. In addition to the usual increase of binder (quicklime + cement) content with decreasing density, it is important to stress that the quicklime / cement ratio varied from 0.61 in the low density recipe (P2 – 0.35) to 1.06 in the high density recipe (P4 – 0.55). This was thought to be representative of the typical European AAC formulations.

Table 3

RETURN SLURRY COMPOSITION FOR THE DIFFERENT AAC RECIPES.

Constituent	Units	P2 – 0.35	P2 – 0.4	P4 – 0.55
Quartz Sand	wt.%	50	55	59
Cement	wt.%	28	25	18
Lime	wt.%	17	15	19
Anhydrite	wt.%	5	5	4
Water/Solid wt. ratio	-	1	1	1

Table 4

USED AAC RECIPES.

	Units	P2 – 0.35	P2 – 0.4	P4 – 0.55
Target density	kg/m <sup>3</sup>	300-350	350-400	500-550
Target compressive strength	MPa	> 2	> 2	> 4
Return Slurry	wt.%	20	20	20
Quartz Sand	wt.%	50	55	59
Cement	wt.%	28	25	18
Lime	wt.%	17	15	19
Anhydrite	wt.%	5	5	4
Aluminium Paste	wt.%	0.22	0.16	0.08
Water/Solid wt. ratio	-	0.68	0.65	0.65

Water was heated to 70°C prior to mix with the other ingredients. This allowed to have a batch temperature close to 40°C at the end of the mixing process.

## 2.2. Methods

### 2.2.1. Raw Materials

Quicklime reactivity was measured according to EN 459-2. As a reminder, t<sub>60</sub> is the time (in min) needed to reach a temperature of 60°C when mixing 150 g of quicklime in 600 g of water initially at 20°C. The higher t<sub>60</sub>, the lower the reactivity. Values for t<sub>60</sub> of the tested quicklimes are listed in Table 2. The full slaking curves are given in Figure 1.

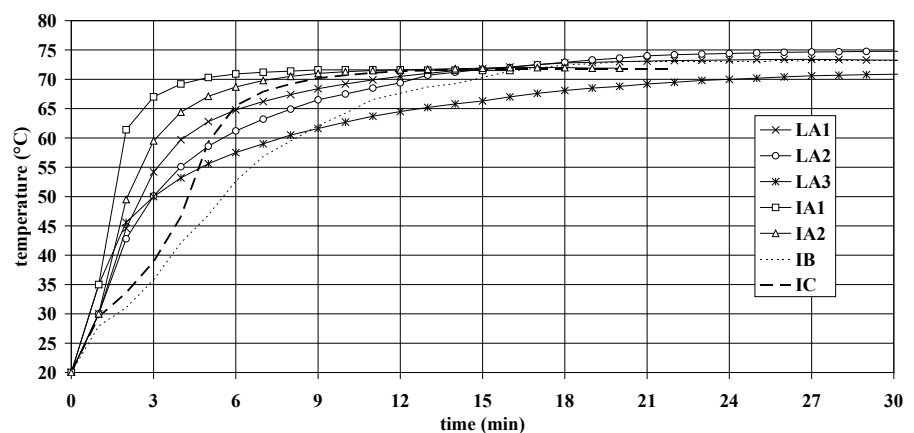


Fig. 1. Slaking curves for all studied quicklimes.

In addition, X-ray fluorescence spectroscopy was used in order to determine the chemical composition of the materials. The data were expressed in terms of equivalent oxide composition and are also listed in Table 1 and Table 2.

### 2.2.2. Green Cake

Green cake was prepared by mixing the different ingredient (Table 4) in a 30-litre bucket with a high speed stirrer at 900 rpm with vertical shaft and disk impeller of 10 cm diameter. The ingredients were added in the following order:

1. Return slurry,
2. Hot water,
3. Quartz sand,
4. Mixture of cement + quicklime + anhydrite (dry mixed prior to incorporation),
5. Aluminium paste.

The overall mixing sequence lasted less than 5 min, and only 20 s of mixing were maintained after having added the aluminium paste.

The suspension was then immediately poured into thermally-insulated 120 x 340 x 280 mm<sup>3</sup> moulds (Figure 2). The mould lid was equipped with a ultrasound sensor and a thermocouple, both connected to a datalogger (Figure 2). This way, both green cake height and temperature were monitored as a function of time in a continuous way.

The green cakes were left to expand for 4 hours. After that time, they were tested for early strength using a pocket-penetrometer equipped with a tailor-made cylindrical testing probe. Early strength was quantified by the penetration value measured in kPa.

Autoclaving was done in a 225-litre autoclave. Moisture-saturated pressure was increased linearly in 2 hours and left at 11 bars for 6 hours. Temperature in the autoclave would typically reach 190°C at 11 bars. The pressure was then released linearly to atmospheric pressure in 2 hours. AAC blocks were then obtained.

### 2.2.3. AAC

6 cubic specimens with dimensions 100 x 100 x 100 mm<sup>3</sup> were sawn from each AAC block, 3 from the top and 3 from the bottom.

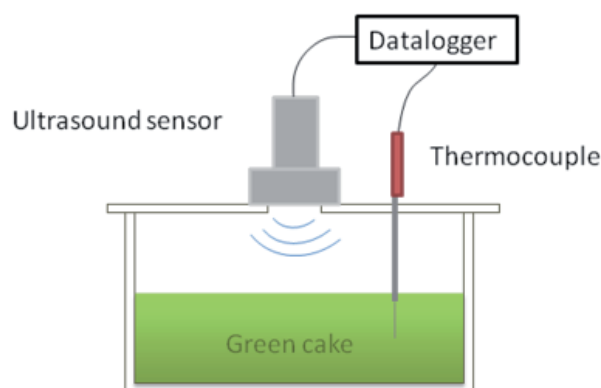


Fig. 2. The moulds used for green cake expansion (left) and the set-up used for monitoring (right).

They were weighted immediately after autoclaving in order to measure moisture content.

They were then oven-dried for 26-32 hours at 80°C in order to obtain the standard moisture content of 4-8 % for compressive strength measurement. The cubes were tested with the force being applied perpendicular to the expansion direction. The result was therefore the mean of the 6 values. No significant difference was observed as a function of cube initial position, showing an overall good homogeneity of the expansion process inside the mould.

After compressive strength measurement, the cubes were left to fully dry in an oven for an additional 24 hours at 105°C in order to measure their final density. The result was therefore the mean of the 6 values. Again, no significant difference was observed as a function of cube initial position, confirming the overall good homogeneity of the expansion process.

## 3. Results

### 3.1. Influence of Quicklime reactivity With Fixed Composition

#### 3.1.1. Green Cake Expansion

The expansion monitoring obtained for the green cakes prepared with quicklimes LA1, LA2, LA3, IA1 and IA2 are given in Figure 3 and Figure 4 for the P2-0.35 recipe. This recipe having the highest amount of binder, the exothermy was maximized and differences among products were therefore magnified. The green cake strength at the end of the expansion process is given in Table 5.

As can be readily seen in Figure 3, the higher the quicklime reactivity (i.e., the lower the  $t_{60}$ ), the faster the green cake expansion. This was consistent with the faster exothermic release observed for the more reactive quicklimes as illustrated in Figure 4.

In the case of IA1 and IA2, the reactivity was so fast that the green cake felt back after 15-20 min, that is after its rapid initial growth (Figure 3). An overshoot of order of 5% of the final height can be readily seen with IA1. This feature almost disappeared for the less reactive materials (LA1 and LA2), with a smoother expansion curve without overshoot.

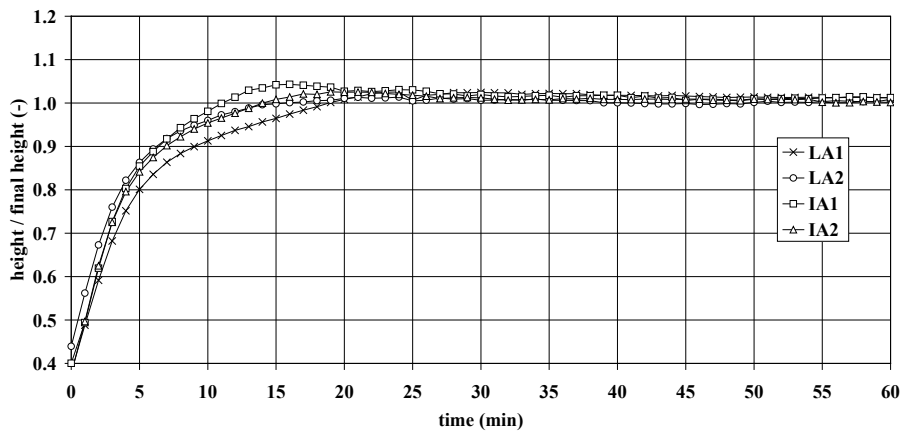


Fig. 3. Green cake expansion (height divided by final height) versus time for the P2 – 0.35 recipes using quicklimes with different reactivity obtained from the same limestone LA1, LA2, IA1 and IA2.

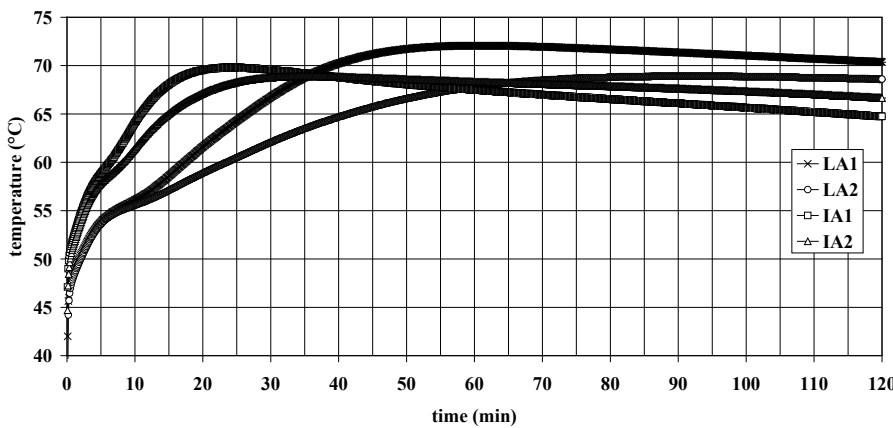


Fig. 4. Green cake temperature versus time for the P2 – 0.35 recipes using quicklimes with different reactivity obtained from the same limestone LA1, LA2, IA1 and IA2. The curves were normalized with the final height.

Table 5

GREEN CAKE STRENGTH (AS MEASURED BY PENETRATION) OBTAINED USING QUICKLIMES WITH DIFFERENT REACTIVITY OBTAINED FROM THE SAME LIMESTONE LA1, LA2, LA3, IA1 AND IA2.

	$t_{60}$	P2 – 0.35	P2 – 0.4	P4 – 0.55
	min	kPa	kPa	kPa
LA1	4.1	1400	2000	4000
LA2	5.2	2000	2200	4800
LA3	7.6	---	3200	---
IA1	1.9	2100	2500	4500
IA2	3.1	1800	2200	5000

Table 6

PROPERTIES OF THE AAC (DENSITY AND COMPRESSIVE STRENGTH  $R_c$ ) OBTAINED USING QUICKLIMES WITH DIFFERENT REACTIVITY OBTAINED FROM THE SAME LIMESTONE LA1, LA2, LA3, IA1 AND IA2. GREY CELLS HIGHLIGHT THE OUT-OF-SPECS DATA.

	$t_{60}$	Density			$R_c$		
		P2 – 0.35	P2 – 0.4	P4 – 0.55	P2 – 0.35	P2 – 0.4	P4 – 0.55
		min	kg/m <sup>3</sup>	kg/m <sup>3</sup>	kg/m <sup>3</sup>	MPa	MPa
LA1	4.1	305	370	499	2.1	3.0	4.7
LA2	5.2	341	346	516	2.1	2.5	5.2
LA3	7.6	---	400	---	---	3.5	---
IA1	1.9	312	378	517	1.8	3.4	5.0
IA2	3.1	302	365	507	2.0	3.0	5.0

In terms of penetration value (Table 5), no significant difference could be observed with the different quicklimes compared at the same density. In fact, penetration values were found to have a  $\pm 300$  kPa range with the same operator. Then, penetration was found to increase with green cake density.

### 3.1.2. AAC properties

The AAC properties for the materials prepared with quicklimes LA1, LA2, LA3, IA1 and IA2 are given in Table 6.

As a general result, target density and target compressive strength could be achieved with almost all materials. Some of the materials were slightly out of the target density range (grey values in Table 6) but modest modifications in the recipe (aluminium paste content especially) would allow to pass the criterion. In all cases where the density was out of the range, the compressive strength was lower but still acceptable, showing that it would be easily corrected by minor modifications of the recipe.

It is interesting that the two reactive quicklimes (IA1 and IA2) that experienced a significant green cake fallback (Figure 3), also experienced a low compressive strength for the low density AAC (P2 – 0.35), to the point that IA1 was even out of specifications (Table 6).

## 3.2. Influence of Quicklime reactivity With VARYING Composition

### 3.2.1. Green Cake Expansion

The expansion monitoring obtained for the green cakes prepared with quicklimes IB and IC, compared to the two extremes from the previous set (IA1 and LA2 with the same  $t_{60}$  as IC) are given in Figure 5 and Figure 6. The green cake strength at the end of the expansion process is given in Table 7.

As can be readily seen in Figure 5, the effect of quicklime reactivity on green cake expansion was less straight forward than before (Figure 3). Quicklimes with very similar reactivities were seen to behave somewhat differently. For example LA2 and IC have the same  $t_{60}$  but IC behaved more like

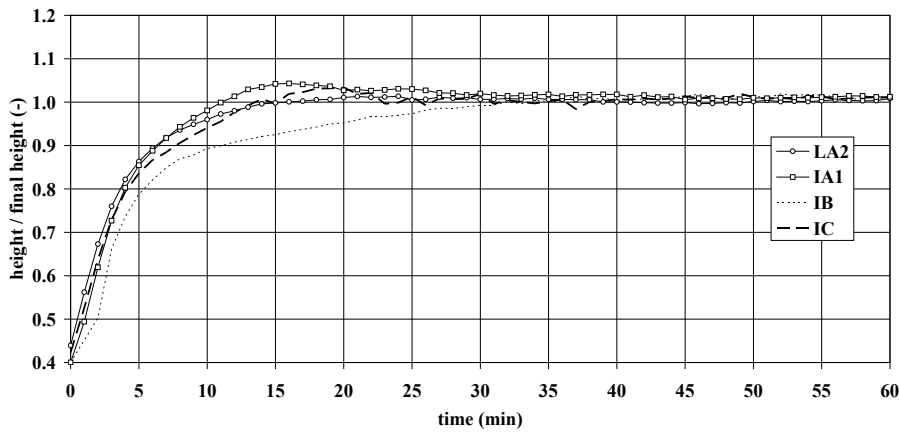


Fig. 5. Green cake expansion (height versus final height) versus time for the P2 – 0.35 recipes with quicklimes IA1, LA2, IB and IC.

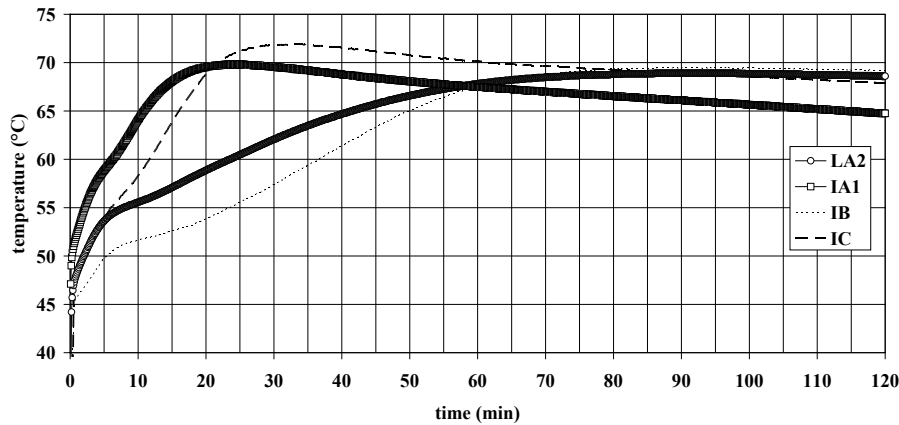


Fig. 6. Green cake temperature versus time for the P2 – 0.35 recipes with quicklimes IA1, LA2, IB and IC.

Table 7

GREEN CAKE STRENGTH (AS MEASURED BY PENETRATION) OBTAINED WITH QUICKLIMES IA1, LA2, LA3, IB AND I.

	$t_{60}$	P2 – 0.35	P2 – 0.4	P4 – 0.55
	min	kPa	kPa	kPa
IA1	1.9	2100	2500	4500
LA2	5.2	2000	2200	4800
LA3	7.6	–	3200	–
IB	8.2	2000	2200	5000
IC	5.2	1800	2500	5000

Table 8

PROPERTIES OF THE AAC (DENSITY AND COMPRESSIVE STRENGTH  $R_c$ ) OBTAINED WITH QUICKLIMES IA1, LA2, LA3, IB AND IC. GREY CELLS HIGHLIGHT THE OUT-OF-SPECS DATA.

	$t_{60}$	Density			$R_c$		
		P2 – 0.35	P2 – 0.4	P4 – 0.55	P2 – 0.35	P2 – 0.4	P4 – 0.55
		min	kg/m <sup>3</sup>	kg/m <sup>3</sup>	kg/m <sup>3</sup>	MPa	MPa
IA1	1.9	312	378	517	1.8	3.4	5.0
LA2	5.2	341	346	516	2.1	2.5	5.2
LA3	7.1	–	450	–	–	3.5	–
IB	8.2	311	379	525	2.3	3.7	5.9
IC	5.2	305	375	522	1.6	2.7	5.1

the highly reactive IA1, with the same impact on the green cake expansion of P2 – 0.35 (Figure 5). Still, green cake expansion kinetics (Figure 5) was always consistent with the kinetics of exothermic release (Figure 6).

In terms of green cake strength (penetration), no difference could be highlighted between the different quicklimes when compared at the same density (Table 7).

### 3.2.2. AAC properties

The AAC properties for the materials prepared with quicklimes IA1, LA2, LA3, IB and IC are given in Table 8.

Interestingly, quicklime IC was found to have a too-low compressive strength with the P2 – 0.35. This behaviour was not observed with quicklime LA2 although it had a similar  $t_{60}$  but was observed with the very reactive IA1 with a much shorter  $t_{60}$ .

## 4. Discussion

From the above results, it appears that quicklime reactivity dramatically affects green cake expansion. When all other parameters are fixed (recipe, modus operandi and limestone origin), the more reactive the quicklime, the faster the expansion rate of the green cake

(Figure 3). This was in agreement with the higher exothermy observed with more reactive quicklimes (Figure 4) and was of course totally expected. Note also that too fast expansion led to incorrect pore structure, with significant bubble coalescence and break-up, as detected by the significant overshoot for quicklimes IA1 and IA2 (Figure 3) which in turn generated a low compressive strength (Table 6). This effect was so pronounced with IA1 that the P2 – 0.35 recipe felt out of the corresponding specification (Table 6). As a result, with the chosen recipes, the chosen modus operandi and this particular limestone origin (A), a  $t_{60} \geq 5.2$  min would have been recommended.

However, use of quicklimes from other origins and reactivities showed that quicklime reactivity, as measured by the slaking curve and quantified by  $t_{60}$ , is not sufficient to anticipate the correct course of AAC production process. Even if IC quicklime had the proper reactivity based on the results

for limestone A ( $t_{60} = 5.2$  min), its effect on green cake expansion was similar to that of the most reactive quicklime IA1 made out of limestone A: The fallback phenomenon was also observed (Figure 5) and the compressive strength of the low density recipe was equally out of specifications (Table 8).

This highlights that quicklime reactivity, as measured by the slaking curve and quantified by  $t_{60}$ , is not sufficient to predict AAC final properties with a given recipe and *modus operandi*. Quicklime origin, and especially the parent limestone source, probably through the quicklime chemical composition, is an important factor controlling AAC properties, especially through its effect on the expansion phase.

In all cases, note that these differences in green cake expansion could probably have been easily corrected by minor changes of the process. In particular, the too-reactive limes (IA1, IA2 and IC) would probably lead to correct AAC at all densities by, for example, only decreasing the initial temperature of the added water.

## 5. Conclusion

The effect of quicklime origin and reactivity on AAC production was studied using both quicklimes from the same limestone but of different reactivities and quicklimes from other limestones and of different reactivities.

For the purpose three recipes were used, with three different target densities corresponding to the European classes P2 – 0.35, P2 – 0.4 and P4 – 0.55.

Green cake expansion was monitored and temperature was recorded continuously during 4 hours for all quicklime / recipe combinations. Green cake strength was measured by penetration at the end of the expansion phase. The blocks were then autoclaved and their compressive strength and density were measured.

From this study and for the tested materials with the given procedure (recipes and methods), it appears that:

- Quicklime reactivity had little effect on green cake penetration value, showing that the density is the main parameter of this property,
- Quicklime reactivity had a major effect on green cake expansion. Quicklime of high reactivity generated a poor pore structure as detected by both the fallback effect and the below-specification compressive strength of the low density AAC,
- Quicklime slaking curve was not sufficient to assure the green cake expansion for a given recipe. Particularly, two quicklimes with similar reactivity as measured by  $t_{60}$  could have very different impact on the production process of AAC, one being too reactive when the other was just right,
- The influence of the too-reactive quicklimes would probably have been easily corrected by decreasing the water temperature.

In all cases, additional work is needed in order to highlight what other quicklime property could allow for the anticipating their influence on the AAC production process, all other parameters (ingredients, recipe, process) being fixed.

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