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Effect of limestone content on properties of portland limestone cement produced by intergrinding

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KEYWORDS

Limestone
Admixture
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Properties
Content

ABSTRACT

Limestone is used very commonly as a mineral admixture in the production of blended Portland cement because of its availability and low cost. Since using limestone reduces the amount of clinker used, it helps to reduce CO_2 emission from the cement production process. Limestone is added into cement commonly by intergrinding with clinker, gypsum and other admixtures (if any). International studies show that the influence of limestone depends not only on the content and fineness of limestone but also on the mineral composition of the clinker used. The objective of this study was to evaluate the influence of limestone content on some properties of Portland limestone cement produced by intergrinding method using materials available in Vietnam. The limestone contents used were 0, 10, 20, and 30%. The properties of the cement investigated include particle size distribution, water of consistency, setting time and compressive strength at ages of 1, 3, 7 and 28 days. The results show that, when increasing the limestone content, the cement particle size was distributed more widely, the water of consistency increased by 1%, and the initial and final setting time decreased slightly. The compressive strength also tended to decrease, the level of decrease was highest at the age of 1 day, gradually decreasing in the later ages. Based on the degree of strength decrease at ages of 3 and 28 days, the limestone content used should not be more than 20%.

1. Introduction

Limestone is commonly used as a natural mineral admixture in the production of blended Portland cement because it is readily available near cement factories and at low cost. Therefore, the use of limestone helps to reduce product cost. In addition, the use of limestone reduces the clinker content in cement, thereby reducing CO_2 emission into the environment from the cement production process.

Results of studies in the world show that the influence of limestone on the properties of cement depends on the fineness and content of limestone added to the cement [1]. Using limestone finer than cement grains, the limestone particles exhibit filling effect that increases the density of the cement matrix structure [2]. Therefore, it can increase the workability, chemical resistance of cement mortar and concrete if using limestone at the right concentration [3]. However, using high amounts of limestone reduces the compressive strength of cement [4]. The fine limestone particles also exhibit nucleation effect on C-S-H crystallization due to their structural similarity, thus they promote the hydration of C_3S [5-7]. Limestone also reacts with C_3A to form calcium carboaluminate hydrates, however, these reactions have little effect on the strength of cement [8].

The degree of influence of limestone on the properties of cement depends on the mineral composition of the Portland cement [9]. Research results of Vuk et al. [9] shows that when using clinker with high lime saturation factor (LSF), limestone mainly affects early strength; while when using clinker with low LSF, limestone mainly affects strength after 28 days and setting time.

Limestone can be added into blended Portland cement in two ways: intergrinding with clinker and gypsum; or grinding it separately and then mixing it with finely ground Portland cement. The intergrinding method has the advantage of being simple, the mill does both grinding and mixing tasks. However, since clinker and limestone have different grindability, the proportion of components will be different in different particle size ranges [10]. The separate grinding method is more complicated, requiring two separate grinding and mixing stages, but it allows easy control of the particle composition of each component, so it is easier to optimize the advantages of limestone. The separate grinding method is usually limited to production of special cements.

In Vietnamese cement factories, limestone is added into blended Portland cement usually by intergrinding method. Although, many studies have been done in the world, but the mineral compositions of clinkers used are different from those of clinkers produced in Vietnam. While there are very few relevant studies published in Vietnam. To be able to use effectively limestone in the production of blended Portland cement in Vietnam, it is necessary to conduct studies on the use of domestic materials.

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The purpose of this study was to examine the influence of limestone content on some properties of Portland limestone cement produced by intergrinding method using materials available in Vietnam. The limestone contents used were 0, 10, 20, and 30%. The properties of the cement studied include particle size distribution, water of consistency, setting time and compressive strength at ages of 1, 3, 7 and 28 days.

2. Materials and experimental procedure

2.1. Materials

The clinker used in this study was produced by VICEM But Son cement company. The chemical and mineral compositions of clinker are shown in **Tables 1** and **2**.

The gypsum used was a natural gypsum whose chemical composition is shown in **Table 3**.

The limestone used was a natural limestone whose chemical composition is shown in **Table 4**.

Table 1. Chemical composition of clinker.

Oxide	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	K_2O	Na_2O
Content (%)	21.36	5.52	3.39	64.73	2.94	0.26	0.73	0.13

Table 2. Mineral composition of clinker.

Mineral	C_3S	C_2S	C_3A	C_4AF	free CaO
Content (%)	47.3	25.8	8.9	10.3	2.7

Table 3. Chemical composition of gypsum.

Composition	SO_3	$CaSO_4.2H_2O$	combined H_2O	residue
Content (%)	45.63	90.20	18.88	1.98

Table 4. Chemical composition of limestone**.**

Oxide	SiO_2	Al_2O_3	Fe_2O_3	CaO	MgO	SO_3	residue	LOI
Content (%)	1.46	0.29	0.13	48.19	4.28	0.04	1.82	43.76

2.2. Experimental procedure

The cement samples were prepared as follows. Clinker, gypsum and limestone were crushed separately by a jaw crusher until all particles passed the 5 mm sieve. Clinker, gypsum and limestone were weighed according to the proportions shown in **Table 5**, then put in a laboratory ball mill and ground until the cement reached the corresponding Blaine fineness. The increase in fineness of cement when increasing the limestone content was to ensure that the sieve residue amounts were ≤ 10% as specified in TCVN 6260:2009 [11].

Table 5. Composition and Blaine fineness of cement samples.

No.	Proportion of components (%)			Blaine fineness
	Clinker	Gypsum	Limestone	(cm^2/g)
1	95	5	0	3600
2	85	5	10	4000
3	75	5	20	4400
4	65	5	30	4800

Blaine fineness of cement samples was determined according to TCVN 4030:2003 [12]. The particle size distribution of cement samples was determined by laser scattering method on a Horiba LA-950 machine. Water of consistency and setting time of cements were determined according to TCVN 6017:2011 [13]. The compressive strengths of cements were determined according to TCVN 6016:2011 [14].

3. Results and discussion

3.1. Particle size distribution

The results of the analysis of the particle size distribution of the cement samples are shown in **Figure 1**.

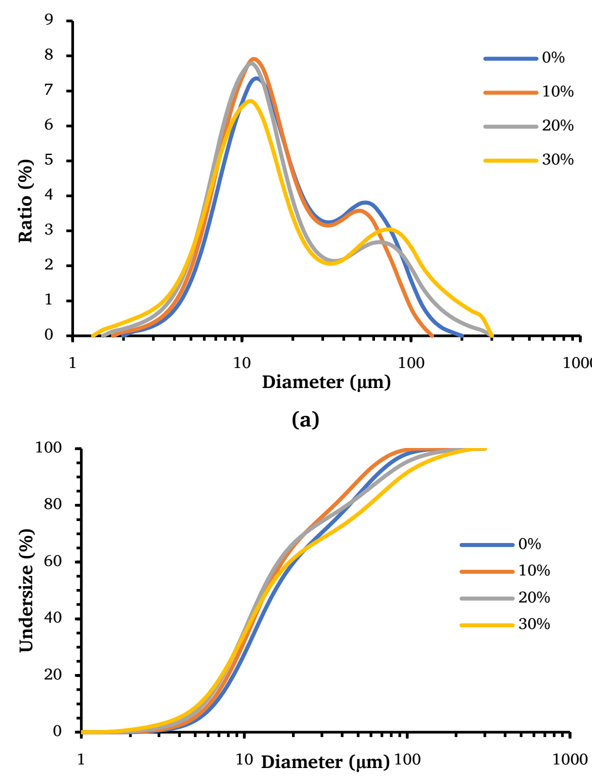

Figure 1. Particle size distribution of cement samples.

The results in **Figure 1a** show that each curve of particle size distribution of the cement samples has 2 distinct peaks, the first peak was higher in the fine-particle region and the second peak was lower in the coarse-particle region. The positions of the first peak of samples were similar, at 11.6 μm, but the positions of the second peak of samples using 0%, 10%, 20%, 30% limestone were different, at 51.5 μm, 51.5 μm, 67.5 μm, 77.3 μm, respectively. This indicates that when the limestone content increased, the particle size distribution of cement was wider. **Figure 1** also shows that when increasing the limestone content, the content of very fine particles and the content of coarse particle increased.

3.2. Water of consistency

The results of water of consistency of cement samples are shown in **Figure 2**. The results in **Figure 2** show that the values of water of consistency of the cement samples containing limestone were higher than that of the sample without limestone. When increasing the limestone content, the water of consistency tended to increase, the increase was 1% when the limestone content increased from 0% to 30%. This increase can be explained by the increase in Blaine fineness (specific surface area) of the cement. Therefore, more water would be required to lubricate the surface of particles for the cement paste to reach the standard consistency [10].

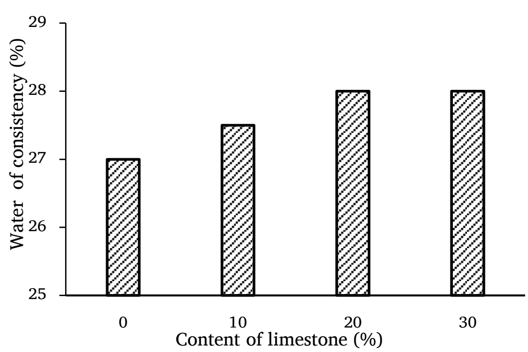

Figure 2. Effect of limestone content on water of consistency of cement.

3.3. Setting time

The experimental results of initial and final setting times of cement samples are shown in **Figure 3**.

The results in **Figure 3** show that, the initial and final setting time of the samples with limestone were shorter than those of the sample without limestone. When increasing the limestone content, the initial and final setting time tended to decrease slightly. When increasing the limestone content from 0% to 30%, the initial setting time decreased by 10 minutes (from 140 minutes to 130 minutes), while the final setting time decreased by 20 minutes (from 245 minutes to 225 minutes). The decrease in setting time would be due to the nucleation effect of limestone on the crystallization of C-S-H hydrate, thus promoting the hydration of C_3S [7]. **Figure 3** also shows that the initial and final setting time of all samples using limestone met the requirements in TCVN 6260:2009 (initial setting time not less than 45 minutes and the final setting time not greater than 420 minutes) [11].

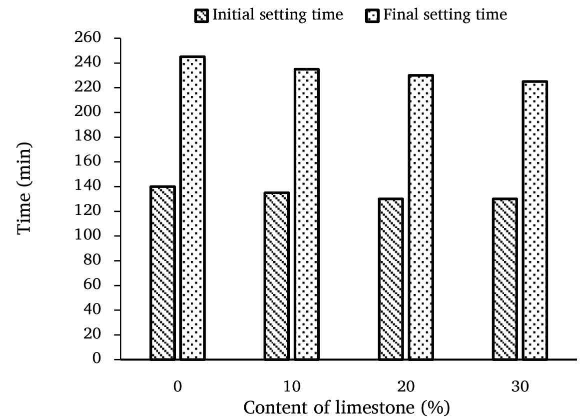

Figure 3. Effect of limestone content on setting time of cement.

3.4. Compressive strength

The results of the compressive strength of cement samples are shown in **Figure 4**.

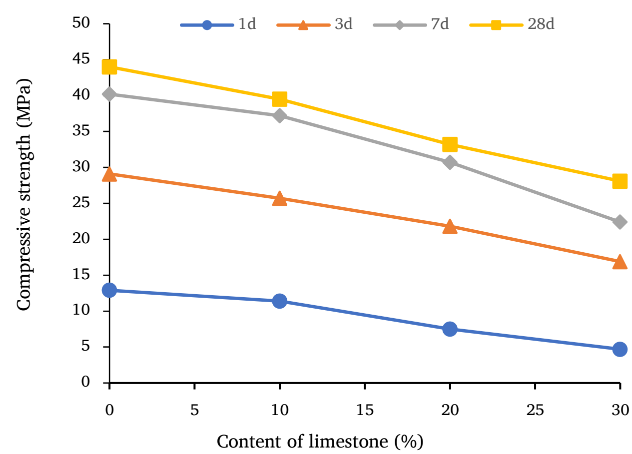

Figure 4. Effect of limestone content on compressive strength of cement.

The results in **Figure 4** show that the samples with limestone have lower compressive strength than the one without limestone. As the limestone content increased, the compressive strength of cement decreased at all ages. The decrease in strength when adding limestone can be explained by the dilution effect of limestone which reduced the content of binder minerals in the cement. As the limestone content increased, this dilution effect increased, leading to further decrease in strength.

Figure 5 shows the degree of decrease in compressive strength of cement samples containing limestone in comparison with the one without limestone. The results show that the degrees of strength decrease when increasing limestone content at different ages were different. The greatest strength decreases were at the age of 1 day when the decreases for samples containing 10%, 20%, and 30% limestone were 12%, 40% and 64%, respectively. The degree of decrease in compressive strength decreased gradually at later ages. For example, at the age of 3 days, the compressive strength of samples containing 10%, 20%, and 30% limestone decreased by 12%, 25% and 42%, respectively. The lowest degrees of strength decrease were at the age of 28 days when the compressive strength of samples containing 10%, 20%, and 30% limestone decreased by only 10%, 24% and 36%, respectively. The change in the degree of decrease in compressive strength at different ages would be due to the change in the limestone content in different particle size ranges of the cement. When increasing the limestone content, the proportion of limestone particles was higher in the fine-particle region and lower in the coarse-particle region; in contrast, the proportion of clinker particles was lower in the fine-particle region and higher in the coarse-particle region. The early strength of cement mainly depends on the content of clinker particles in the fine particle region while the late strength depends on the content of the coarser particles. Therefore, when increasing the limestone content, the lower content of clinker particles in the fine-particle region would result in a greater decrease in early strength; while the higher clinker content in the coarser particle region would reduce the degree of strength loss in late ages.

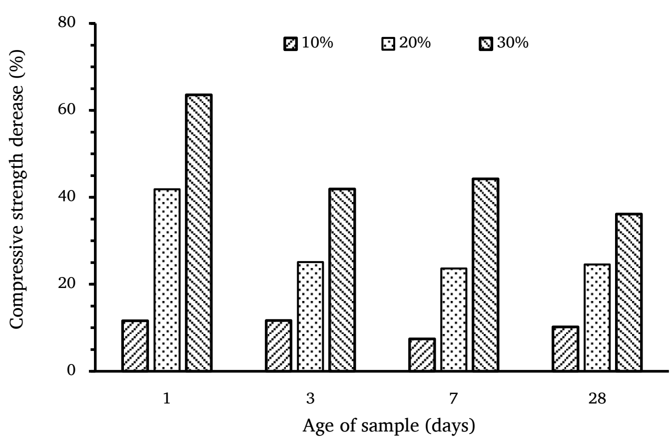

Figure 5. Decrease in compressive strength of cement samples containing limestone in comparison with sample without limestone.

4. Conclusions

From the experimental results of cement samples using limestone content of 0%, 10%, 20%, and 30%, we can draw the following conclusions:

- Increasing the limestone content increased the water of consistency of cement. When the limestone content increased from 0% to 30%, the water of consistency increased by 1%.
- Increasing the limestone content decreased the initial and final setting time of cement. When the limestone content increased from 0% to 30%, initial and final setting time decreased by 10 minutes and 20 minutes, respectively. The initial and final setting time of all samples met the requirements of TCVN 6260:2009 for blended Portland cement.
- Increasing the limestone content decreased the compressive strength of cement at all ages (1, 3, 7, and 28 days). The degree of compressive strength loss at the age of 1 day was greatest, and it tended to decrease at ages of 3, 7, and 28 days.

The standard TCVN 6260:2009 specifies the compressive strength at 3 days and 28 days. Based on the results of degree of compressive strength decrease when increasing the limestone content in this study at 3 days and 28 days old, the limestone content should not be more than 20% to avoid too high decrease in compressive strength of cement. In order to minimize the decrease in compressive strength at the age of 1 day, besides limiting the limestone content to an appropriate level, we might increase the fineness of the cement or use limestone with other admixtures.

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