THE USE OF LIMESTONE POWDER AS AN ALTERNATIVE CEMENT REPLACEMENT MATERIAL: AN EXPERIMENTAL STUDY

Wendimu Gudissa and Abebe Dinku
Department of Civil Engineering
Addis Ababa University

ABSTRACT

This research paper examines the effects of partial substitution of Portland cement clinker with limestone addition on the physical and chemical properties of cement paste and hardened mortar in two ranges of blain fineness values. Laboratory tests were conducted on limestone and clinker samples before used for the intended purpose and then checked for conformity with the Ethiopian and European standards.

The laboratory test results revealed that up to 15% replacement of clinker by fine limestone powder having blain fineness values in the range of 4000 to 4500 cm^2/gm result in comparable compressive strength to similar mixes produced using 100% ordinary Portland cement. It also satisfied the standard compressive strength of high early strength of cement (42.5 MPa) as per EN 197-1 standard requirements. Furthermore, it was found that 25-35% limestone addition by weight results in slightly lesser compressive strength values than the 28^{th} day's standard compressive strength requirement.

Observations on the test results further indicated that increasing in fineness of the limestone filler addition in Portland cement clinker results in relative increase of rate of hydration and faster development of the early age strength while decreasing slightly the consistency and the setting times (initial and final). The loss on ignition is also observed to increase with the increase in percentage proportion of limestone filler addition where as the percentage by mass of sulphate trioxide and insoluble residue as well as soundness effect of the Portland limestone cement remain within the acceptable range.

Key words: *Calcium carbonate, Compressive strength, Early strength, Limestone filler, Loss on ignition, Portland limestone cement*

INTRODUCTION

Production of Portland limestone cement, using limestone filler as a partial replacement to the clinker, is a current trend in the World cement industry; especially in European countries due to its multi-faceted advantages that include: increased cement productivity, reduced production cost and environmental protection with a significant reduction of CO_2 and NO_2 emissions per ton of cement produced.

The recent European Standard EN 197 identifies two types of Portland limestone cements: Type II/A-L containing 6–20% and Type II/B-L containing 21–35% limestone addition. In addition, the inclusion of 5% of filler material that can be calcareous is accepted in all cements. Literatures reveal that, in 2004 the ASTM C150 standard specification for Portland cement was modified to allow the incorporation of up to a 5% mass fraction of limestone in ordinary Portland cements [1].

The substitution of parts of cement by limestone filler have shown several effects on the physical and chemical properties of cement paste and hardened mortar. Most of the research investigations have emphasized that the C_3S hydration rate is accelerated when the amount and fineness of limestone filler ($CaCO_3$) content is increased due to generation of large number of nucleation sites for precipitation of the hydration products [2]. This enhanced degree of hydration at an early age reflected by the improvement of the strength of both mortars and concretes at early ages.

Numerous studies on Portland limestone cement focused on the effects of limestone on the cement performance, participation of limestone in the hydration reactions and the production process specifically intergrinding of clinker and limestone.

The use of limestone filler in combination with Portland cement is a common practice in many countries in the world for various reasons, while this practice has not been exercised in Ethiopia. In recent years, the cost of cements and cement based construction materials in Ethiopia is increasing from time to time and there is a huge gap between demand and supply of cement throughout the country. This rise of cost and demand of cement is mainly due to limited production capacity and restricted types of cement produced in the country for the steady increase in demand of cement for construction.

To minimize this problem and to ensure the continuous development of the construction sector, it is appropriate to increase the production of cement and to look in to production of alternative types of cement that can be used for various construction purposes with relatively lesser cost, technical and ecological benefits.

Portland limestone cement is thus, one of the alternative cement types that can be produced in our cement industries. The raw materials for production of such cement type is sufficiently available, requires lesser energy for production, and has less CO_2 and NO_2 emissions [3]. It is also economical for countries like Ethiopia where OPC and PPC cement types are the only types of cement used for all construction purposes.

This study is thus attempting to make use of these limestone fines in the production of Portland limestone cement. An experimental investigation is thus carried out to examine the impacts of adding these fines on the physical and chemical properties of the cement paste such as consistency, setting time, and compressive strength.

The main objectives of the study described in this paper are, thus to:

i) Study the influence of percentage addition of limestone filler on the physical and chemical properties of Portland cement paste and hardened mortar.

ii) Produce Portland limestone cement as an alternative cement type having comparable characteristics to that of OPC and PPC.

The study further assesses the suitability of the proposed limestone for Portland limestone cements production, and draws conclusions and forward recommendations based on the research findings and indicates areas for future study.

LIMESTONE IN CEMENT PRODUCTION

Limestone is made up of varying proportions of chemicals such as calcium carbonate ($CaCO_3$), Magnesium carbonate ($MgCO_3$), Silica (SiO_2), Alumina (Al_2O_3), Iron oxide (Fe_2O_3), Sulphate (SO_3), and Phosphors (P_2O_5) with calcium and magnesium carbonate being the two major components [4]. Limestone is the most common form of calcium carbonate which is used extensively for the manufacture of cement. Cements in different types are made mainly by calcining a mixture of about 75% limestone and 25% clay to form a calcium silicate clinker which is then ground and mixed with a small amount of gypsum [5].

According to European standard (EN 197) limestone used as a main constituent in cement production shall meet the following requirements [6]:

i) The $CaCO_3$ content shall be ≥ 75% by mass

ii) Clay content methyene blue absorption should be 1.2 g/100 g, and

iii) Total organic material content (TOC) shall be ≤ 0.2% by mass.

Limestone is also one of the well known ingredients used for the production of Masonry cement by intergrinding with Portland cement together with plasticizing material. It provides fattiness and cohesiveness for mortar by improving adherence to the brick or block and preventing bleeding and minimizing water loss due to absorption [7].

THE EFFECTS OF LIMESTONE FILLERS ON CEMENT PROPERTIES

Fineness

Limestone fillers have many effects on the cement properties due to its fineness. Inclusion of this fine material will significantly accelerate the hydration of alite and aluminates of the cement, because the particles act as nucleation sites for the formation of the hydration products. Another effect of finely divided additions is their action as fillers between the cement grains producing a denser paste and densifying the interfacial zone between the aggregate and cement paste [7].

Consistency

The effect of limestone powder on the water requirement of OPC and blended cement has been studied extensively and a majority of findings are in favor of a better workability of mortar and lowering the water requirement for neat paste containing limestone. The improvement in the workability of paste and mortar is due to suitable texture fineness and particle size distribution of cement containing limestone [8].

Soundness

The recent findings show that the addition of calcareous material (limestone) up to the range of 5-7% in cement mortar have smaller influence on shrinkage as compared to siliceous additives. It is also confirmed by different investigators that there is no remarkable effect on the soundness of OPC paste with up to 10% replacement by limestone additives [8].

Hydration

Many research papers on influence of limestone powder on hydration of Portland cement have reported that the C_3S hydration rate is accelerated when the amount and fineness of $CaCO_3$ is increased. This is due to the fact that they generate a large number of nucleation sites for precipitation of the hydration products [8]

Compressive strength

It has been found that addition of limestone powder into cement paste and mortar increases the strength at early ages without changing the workability of mortar. It has also been investigated that blending of Portland cement with 10 - 40% finely ground limestone improves the early strength [9].

Findings of research works on the strength reveals that, irrespective of grinding methods (intergrinding or separate grinding of limestone and other materials) up to 5%, the strength of limestone cement at 3 and 7 days were slightly higher than the pure ordinary Portland cement since increase in strength is directly related to the increase in rate of hydration of cement obtained due to the addition of limestone fillers [9]. However findings also reveal that, as the percent of substitution of limestone in OPC increases, the compressive strength development of resultant cement decreases [9].

Grinding

Studies have shown that intergrinding clinker with different proportion of limestone resulted in improved grinding behavior of clinker resulting in saving of grinding time and decrease in fuel and electric consumption cost. Thus the European cement industries allowed using mineral addition to economize the production of cement under the specified standard. It has also been proved that intergrinding have resulted in better particle size distribution for the same energy level than that of separately grinding of raw materials with clinker [10].

Environmental impact

It has been recognized that cement industries release different gases to the atmosphere including greenhouse gas emissions, Dioxin, NO_2, SO_2, and vibration during operating machinery and blasting in quarries. It is an established fact that 0.9 ton of CO_2 is emitted per ton of cement [11]. However, there are strategies for the reduction of carbon dioxide which aimed at lowering emissions per ton of cement, even though there is inherent emission of carbon dioxide during chemical breakdown of the limestone in cement kilns during production of Portland cement clinker.

One of the strategies of decreasing CO_2 emission is intergrinding or blending limestone with Portland cement which offers key advantages in reduction of CO_2 emissions, climate change, economic and technical benefits [10].

PROPERTIES OF MATERIALS USED IN THE INVESTIGATION

Clinker

The clinker used for this investigation was Portland cement clinker produced by Mugher cement factory. The suitability of this clinker was investigated from its chemical and mineralogical compositions which are the major properties responsible for the quality of cement.

To this effect, the clinker chemical and mineralogical compositions were tested in Mugher cement enterprise materials quality testing and assurance laboratory and the test results are shown in Table 1 and 2, respectively.

Table 1: Chemical compositions of Mugher Portland cement clinker

Chemical composition	Mugher clinker (%)
SiO_2	22.15
Fe_2O_3	3.43
Al_2O_3	5.76
CaO	65.05
MgO	1.05
SO_3	1.04
LOI (Loss on Ignition)	0.08
IR (Insoluble residue)	0.05
F-CaO (Free calcium)	0.67
SM (silica Modules)	2.41
AM (Aluminum Modulus)	1.68
100LSF (Lime Saturation Factor)	91.93
CI (Coating Index)	32.35
LPH (Liquid phase)	26.19
$CaCO_3$	76.39

Table 2: Mineralogical compositions of Mugher cement clinker

Mineralogical composition	Percent (%)
C_3S	47.11

C_2S	27.96
C_3A	9.46
C_4AF	10.44

The above mentioned clinker was produced from over burden limestone, clay and sand stone which were collected from the nearby raw materials sources of the factory located 2.5, 7.5 and 5km distances from the plant site, respectively.

Limestone filler

The limestone filler used in the test program was obtained from the source of Mugher limestone raw material exploration site which is 2.7 km away from the factory. While selecting the sources, the potential for being used in the cement industry was also taken into consideration. The European standard EN 197, for limestone to be used as additives or as main constituent filler in cement requirements was also considered.

Laboratory test results indicated that, the $CaCO_3$ content was 93.75% which is greater than the standard requirement (75%) as given by EN 197 and the clay content, was found to be 1.15g/100g which is also below the standard requirement (1.2 g/100g).

CEN Standard Sand

The sand used for the study to determine the strength of cement was CEN standard sand which is well graded rounded particles having a silica content of 98 % as specified in EN 196-1 standard requirements. This CEN standard sand is delivered in plastic bags with a content of 1350 g.

Water

Throughout the investigation, tap water supplied for drinking consumption at Mugher were used for curing the hardened mortar samples and also distilled water were used for all physical and chemical analysis works.

Chemicals

For the determination of the chemical and mineralogical composition as well as sulphate and insoluble residue, different chemicals were used as per the specified method of testing cement based on the European standard EN 196-2.

EXPERIMENTAL PROGRAM

The experimental program was composed of the following major sections:

i) Determination of the effects of fineness and percentage additions of limestone fillers on various properties of Mugher Portland cement clinker, and

ii) Study of the effects of fineness ranges and percentage additions of limestone fillers on Mugher Portland cements to determine the contents of the chemical composition.

A total of 18 mix proportions (Table 3) were designed to study all parameters in addition to the control mix made without limestone fines. Limestone fines were added in different percentages ranging from 5 to 35% with 5% increments and two blain values varying between 4000 - 4300 cm^2 and 4301 - 4500 cm^2/gm.

Table 3: Mix proportion of cement with blain fineness values of 4000 - 4300 cm^2/gm and 4301 - 4500 cm^2/gm

Mix Code	Mugher clinker (%)	Pumice (%)	Gypsum (%)	Limestone Fillers (%)	Blain fineness (cm^2/gm)	Remark
M1	95	-	5	0	4000-4300	Control mix
M1	70	25	5	0	4000-4300	Control mix
M1-05	90	-	5	5	4000-4300	
M1-10	85	-	5	10	4000-4300	
M1-15	80	-	5	15	4000-4300	
M1-20	75	-	5	20	4000-4300	
M1-25	70	-	5	25	4000-4300	
M1-30	65	-	5	30	4000-4300	
M1-35	60	-	5	35	4000-4300	
M2	95	-	5	0	4301-4500	Control mix
M2	70	25	5	0	4301-4500	Control mix
M2-05	90	-	5	5	4301-4500	
M2-10	85	-	5	10	4301-4500	
M2-15	80	-	5	15	4301-4500	
M2-20	75	-	5	20	4301-4500	
M2-25	70	-	5	25	4301-4500	
M2-30	65	-	5	30	4301-4500	

M2-35	60	-	5	35	4301-4500	

Mix proportion

The following test programs were followed in the investigation.

The 1st series of mixes (2 in number) were prepared as a control mix in order to compare the physical and chemical properties of cement using 100% Mugher ordinary Portland cement and 100% Mugher Portland Pozzolana cement with no limestone filler addition as shown in Table 4 below. The 2nd and 3rd series of mixes (7 in number each) as indicated in Table 4 below were prepared to compare the chemical and physical properties of cement.

Limestone filler additions were then added with a weight increment of 5% varying from 5% to 35 %. In these mixes the fineness of the cement used for conducting the test were with cement blain values ranging from 4000 - 4500 cm^2/gm. Detail mix proportion data and other relevant information are reported elsewhere [12].

Table 4: Laboratory sample cement preparation data

Mix	Mix Code	Clinker (gm)	Gypsum (gm)	Pumice (gm)	Limestone Filler (gm)
Mix I	M1	2850	150	0	0
	M1	2100	150	750	0
	M2	2850	150	0	0
	M2	2100	150	750	0
Mix II	M1-05	2700	150	-	150
	M1-10	2550	150	-	300
	M1-15	2400	150	-	450
	M1-20	2250	150	-	600
	M1-25	2100	150	-	750
	M1-30	1950	150	-	900
	M1-35	1800	150	-	1050
Mix III	M2-05	2700	150	-	150
	M2-10	2550	150	-	300
	M2-15	2400	150	-	450
	M2-20	2250	150	-	600
	M2-25	2100	150	-	750
	M2-30	1950	150	-	900
	M2-35	1800	150	-	1050

***Grinding* of the raw materials**

In the preparation of laboratory samples, a laboratory ball mill having 420 mm diameter as shown in Figure 1 below, was used for grinding processes. The revolution rate for the grinder was about 45 revolutions per minute. The grinding elements used were balls of different diameters. The size distributions of the grinding elements used are given in Table 5. The grinding elements weight and distribution filled into the ball mill was identical in all grinding operations.

Figure 1 Grinding mill used for grinding and preparing cement (Photo from Mugher Cement Factory Laboratory).

Table 5: Size and distribution of grinding elements

Spherical Balls diameter (mm)	No. of balls (pc)	Mass of each ball (kg)	Total mass of ball (kg)
40	22	0.35	7.7
50	12	0.8	9.6
80	4	2.1	8.4
Total			25.7

Specimen preparation

For all mixes the European standard sand with particle size distribution and moisture content complying with the specified standard of EN 196-1: 1994 were used. The specimens for strength tests were then cast in steel moulds (40 mm x 40 mm x 160 mm) in two layers and compacted on jolting apparatus with 60 jolts for the first layer and then compacted after the second layer with a further 60 jolts. All specimens were prepared with one part cement to three parts of standard sand proportioned by weight with the addition of water. All specimens were cured in curing pond for either 2 or 28 days.

EXPERIMENTAL RESULTS AND DISCUSSION

Suitability of limestone fillers as additives for the production of Portland limestone cement

Laboratory test results indicated that (Table 6), the calcium carbonate content of the limestone filler is above 93% and its clay content is 1.15gm/100gm satisfying EN 197-1 standard requirements. Furthermore, the test results also fulfill the requirements of researchers which determine the chemical composition of limestone for favorable utilization as additive in Portland cement to be with $CaCO_3$ > 9O %, Al_2O_3 < 2 %, MgO < 5% and SO_3 < 0.5 [8].

Table 6: Laboratory test results on chemical composition of limestone powder samples

Chemical composition	Norm used by the Factory (%)	Test result of Limestone Powder (%)
$CaCO_3$	≥ 90.0	93.75
SiO_2	≤ 5.0	2.58
Fe_2O_3	≤ 0.80	0.12
Al_2O_3	≤ 1.50	1.03
CaO	≥ 50.0	53.16
MgO	≤ 0.80	0.4
SO_3	≤ 0.30	Negligible
K_2O	≤ 0.30	Negligible
Na_2O	≤ 0.30	Negligible
LOI	≥ 40.0	41.76

Comparing the test results shown in Table 6 with EN 197-1 standard, and with past investigators report, the limestone raw material quarried from Mugher cement factory source is suitable for utilization as addition to cement clinker in the production of Portland limestone cement under controlled and specified cement quality standards.

Grinding time

It is observed from the test results that for the same range of blain fineness value the water requirement of MPPC is higher than that of MOPC and limestone added cement. This increase of water requirement is due to increase of surface area of cement particles of MPPC, a slightly lower hydration reaction of MPPC to that of MOPC and limestone added cements. However, for the 5% addition of limestone fillers the water requirement doesn't show significant difference comparing to MOPC.

As expected, more time is required for fine grinding than normal fineness. However, the addition of limestone filler reduces the grinding time and also decreases energy consumption of the mills as compared to the production of the same cement quality of pure Portland cement of the same fineness. This decrease in grinding time is due to softness of limestone and easiness to grind than pure Portland cement clinker.

Consistency of cement paste

Normal consistency tests were conducted to observe the changes in water requirements of pastes due to limestone additions. The test results reveal that the consistency of cement pastes slightly decreases with increase of blain fineness values and with increased proportion of limestone content of the cement (Table 7). This decrease in consistency is due to the addition of limestone which increases the plasticity of cement paste. This may be attributed to the effect of limestone as an active component in the hydration of Portland cement, i.e. the rate of hydration increases and the amount of the hydration products enhances. The limestone fillers act as a nucleating agent which increases the hydration rate of cement paste. And also the limestone forms monocarbo aluminate hydrate that needs less water than that of ettringite [9].

Table 7: Consistency of the cement paste as a function of percentage substitution of limestone

Mix no.	Mix Code	Consistency (%)	
		4000 - 4300 cm^2/gm	4301 - 4500 cm^2/gm
1	M1	26	26
2	M2	32	28
3	M-05	25	24
4	M-10	24	24
5	M-15	23	23
6	M-20	23	23
7	M-25	23	22
8	M-30	23	22
9	M-35	22	22

Setting time

The EN 197-1:2000 limits the initial setting times for composite Portland cement not to be less than 45 minutes.

Ethiopian standards also specifies initial and final setting time for Portland Pozzolana cement (ES C.D5.202, Section 4.2.4) to be 45 minutes and 600 minutes, respectively. Comparing the obtained test results of investigation indicated in Table 8, all limestone added cement produced satisfy the requirements specified by both European and Ethiopian standards.

The test results also indicate that, limestone addition into Portland cement increases the initial and final setting times considerably compared to Mugher OPC due to dilution of C_3S and C_3A content in the cement. However, as indicated in Table 8 Mugher PPC shows slightly higher setting time compared to limestone added cement.

As expected, it is also noticed that as the fineness increases from Blain fineness value ranging from 4000-4300 cm^2/gm to Blain fineness value range of 4300-4500 cm^2/gm, the setting time of the limestone added cement is slightly accelerated.

Soundness

Le-Chatlier apparatus was used to conduct soundness test. No remarkable effects are observed on the soundness for replacement of cement clinker up to 35% by limestone additives.

This negligible soundness effect can be attributed to less content of MgO and free CaO in the raw materials, fineness of the cement, good chemical and mineralogical composition of the clinker and good burning temperature in a kiln which favor a decrease of free periclase to occur.

Table 8: Initial setting time of the limestone added cement paste

No. of mix	Mix code	Initial Setting time (minute)		Final Setting time (minute)	
		4000-4300 cm^2/gm)	4301-4500 cm^2/gm	4000-4300 cm^2/gm	4301-4500 cm^2/gm
1	M1	117	84	165	137
2	M2	140	144	187	220
3	M-05	125	101	178	148
4	M-10	127	109	182	158
5	M-15	128	112	184	162
6	M-20	130	115	185	168
7	M-25	131	119	192	168
8	M-30	135	125	193	168
9	M-35	145	136	197	172

Loss on Ignition, Sulphate and Insoluble Residue

The European standard EN 197 has defined limestone Portland cement as a separate cement type and as a standard product under EN 197-1 with two classes, one with addition of 6-20% (II/A-L) and another with 21-35% (II/B-L) limestone additions [6]. However the standard requirements for the total loss on ignition, sulphur trioxide and insoluble residue for the Portland limestone cement products are not specified as the other composite Portland cement in EN 197-1.

Table 9: Test results of loss on ignition, sulfur trioxide and insoluble residue of cement with Blain fineness value from 4000 - 4300 cm^2/gm.

No. of mix	Mix code	Cement with Blain fineness value of 4000- 4300 cm^2/gm			Cement with Blain fineness value of 4301- 4500 cm^2/gm		
		Sulphate, SO_3 (%)	Loss on Ignition, LOI (%)	Insoluble Residue, IR (%)	Sulphate SO_3 (%)	Loss on Ignition, LOI (%)	Insoluble Residue, IR (%)
1	M1	2.50	1.37	2.00	2.88	1.32	1.37
2	M2	2.77	3.42	22.6	2.57	4.36	20.88
3	M-05	2.70	3.13	1.68	2.90	3.76	0.75
4	M-10	2.70	5.33	1.87	2.85	5.78	1.36
5	M-15	2.78	7.46	1.63	2.60	8.35	1.44
6	M-20	2.54	9.30	1.73	2.59	9.80	3.71
7	M-25	2.84	11.59	1.64	2.31	11.91	2.93
8	M-30	2.64	13.69	2.03	2.72	14.11	3.35
9	M-35	2.72	15.65	1.88	2.58	15.89	5.01

However as shown in Table 9 above the sulphate (SO_3) content of limestone added cement is within the specified standard of EN 197-1 Composite Portland Cement and does not significantly vary as limestone filler content addition and fineness increase.

As it is also noted from the investigation, an increase in insoluble residues is observed with increase of limestone content which doesn't exceed the specified percentage stated in European standard. The increase in fineness of the cement also doesn't show significant difference with respect to insoluble residue. It is, therefore, possible to conclude that the material used for the production of Portland limestone cement were free from impurities or adulterants largely arising from gypsum and added limestone.

However, for both Portland composite cement and Portland limestone cement the specification on European standard does not specify the upper and lower limit of insoluble residue as other cement types. This calls for attention when using these two types of cement in order to avoid any unnecessary reaction that may occur with cement after hardening of mortar and concrete in areas of reactive environment.

Higher values in loss on ignition are observed as shown in Table 9 above due to replacement of clinker by limestone at different progressive percentage and also due to the existence of water of hydration in the gypsum.

From different research works cement with limestone addition or filler in to the cement will have relatively high LOI values as a result of the decomposition of $CaCO_3$ [8]. Maximum loss on ignition permitted by BS 12:1991 and ASTM C150-94 is 3% and 4% respectively. But with cement containing calcareous filler, 5% of the mass of the cement nucleus is allowed by EN 197- 1992.

Even though the upper limit for loss on ignition ensures cement freshness, there is no specified standard requirement set for loss on ignition for Portland limestone cement on the EN 197-1, special consideration thus need to be taken during utilization for the intended purpose of work.

Compressive Strength of Hardened Mortars

Mortar specimens were prepared in accordance with methods of testing cement EN 196-1 to determine strength tests. Tests were made with limestone added cements and Portland cements for determining 2 and 28 days compressive strengths. The summaries of mean strength test results obtained are shown in Table 10.

The test results indicate that with the exception of 35% addition, which shows slightly less compressive strength, all the remaining limestone added cement satisfy the 2 days compressive strength requirement of EN 197-1.

As it is also indicated in Tables 10, the test result of the 28th day compressive strength of hardened mortar, addition of limestone filler from 5% to 10% and 5% to 15% with the Blain fineness values of 4000-4300 cm^2/gm and 4301 – 4500 cm^2/gm respectively, satisfies the quality of high early compressive strength of cement of 42.5 MPa and comply to European standard requirements.

Table 10: Summary of mean compressive strengths of mortar samples [MPa]

No. of mix	Mix code	Cement with Blain fineness value of 4000- 4300 cm^2/gm		Cement with Blain fineness value of 4301- 4500 cm^2/gm	
		2 days	28 days	2days	28days
1	M1	30.64	61.90	33.01	63.07
2	M2	26.33	48.48	28.09	56.49
3	M-05	22.93	45.27	24.75	52.87
4	M-10	19.16	41.09	20.73	44.57
5	M-15	15.61	35.35	16.37	37.39
6	M-20	12.47	29.62	12.65	30.57
7	M-25	10.00	25.75	11.26	34.00
8	M-30	8.99	29.90	10.04	26.85
9	M-35	6.75	18.10	7.82	22.80

As the test results indicate, compressive strength of all hardened cement pastes increase with increase of age. The incorporation of fines having a high specific area from 4300-4500 cm^2/gm also improves compressive strengths considerably when compared to 4000-4300 cm^2/gm at 2 and 28 days. This increment of strength is due to the increase of fineness of cement which has direct relationship with the strength development.

It can also be observed that the compressive strength of hardened cement mortar decreases simultaneously with the increase in addition of limestone filler content for same blain fineness. These decreases in strength mainly occur due to replacement of Portland cement clinker with limestone addition with different proportion causing dilution of C_3S and C_2S composition of Portland cement which are responsible for the strength development.

Limestone as an addition to Portland cement for masonry cement production

Test results indicated that limestone addition to Portland cement clinker beyond 25% shows lower compressive strength at 28^{th} days. These strength values do not fit to the accepted level of the specified European standard requirement.

However, as per the ASTM C-91 specification and EN 413–1: 2004 the compressive strength required for masonry cement is not very high as compared to the cement used for structural works. Masonry cements, however, need air entraining agent for the requirements of workability, setting time, plasticity and water retention and also for ensuring uniform characteristics of masonry cements.

Limestone added cement with higher addition of limestone is found to have lower compressive strength at the age of 28 days. But this higher addition of limestone is appropriate for utilization as masonry cement since very high compressive strength is not normally required for making mortars.

In addition from the research work, limestone added cements gain high rate of early strength due to filler effect and develop early strength quickly which is particularly advantageous during cold weather and also ensures maximum durability from an early age [19].

From compressive strength test results, limestone addition more than 25% with less blain fineness value (less than 4000 cm^2/gm) to Portland cement together with air entraining agent can be used as masonry cement. Since for masonry cement very high compressive strength is not always compulsory for making mortars. But further detail study need to be conducted for utilization of such cement as masonry cement.

CONCLUSIONS AND RECOMMENDATIONS

CONCLUSIONS

1. Limestone raw material around Mugher cement factory used for the investigation satisfy the standard requirement of EN 197-1 for production of Portland limestone cement.

2. The investigation has revealed that replacement of ordinary Portland cement by fine limestone powder from 5% to 10% with Blain fineness value of 4000 to 4500 cm^2/gm satisfies the standard compressive strength requirement of high early strength cement (CEN 42.5) as per the standard requirements of EN 197-1.

3. Replacement of Portland cement clinker by limestone filler from 15% - 20% with Blain fineness value of 4000 to 4500 cm^2/gm satisfies compressive strength of ordinary early strength of 32.5 MPa as per EN 197-1 standard requirement.

   Addition of limestone by weight to the clinker from 25% to 35% results in slightly lesser 28^{th} day's standard compressive strength. But the cement can be used as masonry cement with the addition of air entraining agent.

4. The grinding results indicated that, as the replacement of limestone increases by weight, increases in cement fineness and decrease in grinding time were observed compared to pure ordinary Portland cement. Since limestone is softer to grind than pure clinkers the energy required is also relatively less than required to grind pure clinker for Portland cement production.

5. Portland limestone cements produced by 25% limestone addition obtain better early compressive strength values than that of MPPC for the same fineness values.

6. The addition of limestone filler in to Portland cement clinker results in increase in cement fineness and this fineness of the cement provide higher rate of hydration and hence faster development of the early strength.

7. All prepared cement samples with limestone fillers satisfy the setting time, Le-Chatliers expansion or soundness, sulphate and insoluble residue requirements given in EN 197-1.

8. The rate of compressive strength development of limestone added cements is higher than that of MPPC. However, the rate of strength development decreases with the increase of limestone filler addition by over 25% replacement.

9. The test results indicated that, 28^{th} days compressive and flexural strengths of hardened cement mortar decrease with the increase in the percentage addition of limestone content for same blain fineness and also increase with the increase of fineness.

10. Total Loss on Ignition increases with increase in addition of limestone content in the cement, this increase may be due to decomposition of $CaCO_3$ in to CaO and CO_2.

11. The manufacture of Portland limestone cement leads to reduced levels of CO_2 and NO_2 emissions and reduced energy consumption when compared with the production of the same quantity of pure Portland cement, thus contributing towards sustainability of construction works and better environmental conservation.

RECOMMENDATIONS

Based on the laboratory investigations, the following recommendations are forwarded.

1. Portland limestone cement (limestone added Portland cement) is one of the alternative cementing materials well suited to our construction industry with technical, economical and environmental benefits. Therefore concerned bodies should be made aware and promote production and use of these cement type for appropriate functions.

2. Since there is abundant quantity of limestone with necessary chemical composition, it is possible to produce Portland limestone cement at Mugher cement factory.

3. In Ethiopian construction industry, little is known on availability of different alternative cement types except OPC and PPC. Therefore, cement production companies as well as political decision makers together with professionals particularly material engineers should take an active part to exploit the existing resource for using it economically.

4. Some basic physical and chemical properties of limestone added cements were investigated. However, further investigation can be made on the following topics:

   a) Effects of different fineness values and compositions of limestone fines on Portland cements

   b) Study the durability characteristics of the produced cements such as sulfate resistance and reactivity with alkali-reactive aggregates.

   c) Properties of fresh and hardened concrete prepared with limestone added Portland cements.

   d) Cost benefit analysis of Portland limestone cements.

ACKNOWLEDGEMENTS

The authors of this article would like to thank Addis Ababa University for financing the research work. Many individuals who have kindly contributed in the research work by providing access to important documents and sharing their professional view deserve many thanks. The authors would also like to thank Mugher cement enterprise for their support in providing the laboratory facilities and required research materials.

REFERENCES

[1] *Optimal Criteria of Algerian Blended Cement using Limestone Fines Journal of Civil Engineering and Management* http://www.jcem.vgtu.lt/upload/civil_zurn/269-275-jcem-2008-4-guemmadi.pdf.

[2] Boubitsas, D., *Replacement of Cement by Limestone Filler*: The Effect on Strength and Chloride Migration in Cement Mortars", Nordic Concrete Research No.32,2004, pp.31-44.

[3] Mc Grath, R. J., *The Canadian Cement Industry and Innovation Towards Sustainable Development*, Cement Association of Canada, June 10-13, 2008.

[4] Harris, P. M., *Limestone and Dolomite Minerals*; Mineral Resources Consultative Committee Mineral dossier, No 23, 1982, London.

[5] Abebe Dinku, *The Influence of Limestone Aggregates and Limestone Fillers on the Properties of Concrete*, M.Phil. Thesis, University of Leeds, April, 1991.

[6] EN 197-1, Cement–Part 1: *Composition, Specifications and Conformity Criteria for Common Cements*, European standard, 2000.

[7] Neville, A.M., *Properties of Concrete*, ELBS with Longman, 4th edition, 1996.

[8] Peter C. Taylor, *Specifications and Protocols for Acceptance Tests on Processing Additions in Cement Manufacturing*, National Co-operative High Way Research Program, NCHRP report 607, http://trb.org/publications/nchrp/nchrp_rrd_270. pdf.

[9] Newman, John Brian and Choo, B. S., *Advanced Concrete Technology*: Constituent materials, 2003.

[10] *Use of Perlite as a Pozzolanic addition in Blended Cement Production*, http://web.cemetu.edu.tr/divisions/pages/view_thesis.php?id=693, August 2004.

[11] EN 196-6, *Methods of Testing Cement* – Part 6: Determination of Fineness, European Standard, 1989.

[12] Wendimu Gudissa, *Study on the Use of Limestone Powder as Alternative Cement Replacement Material*, M.Sc Thesis, Addis Ababa University School of Graduate Studies, September 2009.

[13] EN 196-1, *Methods of Testing Cement* – Part 1: Determination of Strength, European Standard, 1994.

[14] EN 196-2, 1995, *Method of Testing Cement*, Part 2 Chemical Analysis of Cement.

[15] EN 196-3, *Methods of Testing Cement* – Part 3: Determination of Setting Time and Soundness, European Standard, 1994.

[16] EN 196-7, 1992, *Method of Testing Cement*, Part 7: Method of Taking and Preparing Samples Cement.

[17] Mindess, S., Young, J. E. and Darwin, D.; *Concrete*, 2nd edition, 2003.

[18] Industrial Mineral Laboratory Manual, Technical Report WG/92/29 Mineralogy and Petrology Series, British Geological Survey, http://www.bgs.ac.uk/research/international/dfid-kar/WG92029 _col. pdf.

[19] Santhkumar, A. R., *Concrete Technology*, 1st edition, Oxford University, 2007.

[20] Bnerjea, H. N., *Technology of Portland Cement and Blended Cements* 2nd edition, published in March 31, 1998, by Ah Wheeler Publishing Co Ltd.

[21] Ghosh, **S.N.**, *Advance in Cement Technology*, Chemistry, Manufacture and Testing, 2nd edition, Published by Tech Books International, New Delhi -110019 India, 2002.

[22] Lundgren, M., "*Limestone Filler as Addition in Cement Mortars*: Influence on Early-Age Strength Development at Low Temperature." Nordic Concrete Research, No.31, 2004, pp 50-63.