

Utilization of Stone Cutting Industry Slurry in Cement Production

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Abstract The research presents a study on the possibility of using natural stone saw powder in the West Bank, which is considered an environmental problem and brings agricultural land pollution. This study examines the possibility of using the powder waste in the Portland lime cement industry to reduce the cost as well as the environmental damage resulting from the cement industry, where different proportions of powder were added on clinker and the effect on the physical and mechanical properties was studied. The results demonstrate that the compressive strength of different mixtures after 28 days slightly decreased upon the addition of 5 wt %, 10 wt. % and 15 wt. % limestone powder in comparison with reference sample without powder, though strength of mixtures complies with standard (EN-197-1-2011) limits for the limestone cement, the percentage of consistency of the prepared samples decreased from 29.5% to 29.0% upon the addition of limestone powder, while both of the initial and final setting times of the prepared samples decreased with the addition of limestone powder, the initial setting time decreased around 35 minutes while the final setting time decreased 30 minutes the addition of limestone powders up to 15 wt. %. The adding powder did not affect negatively on the expansion value of the cement samples, and it had a value of 1.0 mm for all tested samples, which is an acceptable value according to EN-197-1-2011, and the results obtained mean the possibility of using stone saw powder in a positive way in the manufacture of Portland lime cement, thus solving the environmental problem resulting from the stone industry, as well as reducing the

cost of cement and the damage of its manufacture on the environment resulting from the stone industry, as well as reducing the cost of cement and the damage of its manufacture on the environment (global warming).

Keywords Cement, Environmental Benefits, Limestone Slurry Waste, Limestone Cement, Clinker

1. Introduction

The cement industry is considered as one of the strategic industries in all countries due to its importance in construction and reconstruction fields. The international production magnitude of cement reaches nearly 1300 million tons annually [1]. The size of demand of cement in Palestine increases because the Palestinian society passes in construction stage, the demand of cement in Palestine was about 2.7 million tons in 2014 and it was expected to increase in the future [2]. According to the Palestinian National Information Center, the Palestinian market imports all its needs of cement from different countries such as Israel (80%), Jordan (9%), Europe (6%) and Egypt (5%) [1].

Nowadays, work is underway to establish the first mill for the cement industry in Palestine to reduce the amount of imported cement. Therefore, it is necessary to work on providing local raw materials of high quality and low cost, as well as being environmentally friendly. This is what was

studied in this research through the use of stone slurry powder in the production of Portland limestone cement, which is used in construction and finishing works in Palestine.

1.1. Environmental Problems

Producing one ton of cement produces the equivalent of 800 kg of carbon dioxide, which is considered a large number if we know that the world's production is more than 1300 million tons annually, and this means that the cement industry is responsible for approximately 5-8% of global emissions of carbon dioxide [3]. Due to the high energy and fuel requirements of the cement production process, supplemental cementitious materials such as pozzolana, limestone, and slag are now commonly used in cement. Between 1990 and 2020, the consumption of materials will have expanded from 10% to around 50%.

However, Palestine's primary industry is the manufacturing of natural stone, which begins with the extraction of rocks from quarries located throughout the West Bank and their transportation to stone-cutter facilities. Metal saws are used in stone cutting facilities to cut and shape stone blocks, and they require a lot of cooling water to do so. As depicted in Figure 1, the cooling water is released from the plant as a highly viscous substance known as stone slurry waste.

The discarded stone slurry presents considerable environmental problems. This technique degrades the quality of the air after it has dried, destroys the soil's fertility, contaminates the earth, decreases ground water recharge, and exacerbates the drainage issue [4, 5].



Figure 1. Stone cutting industry wastes disposed of in agricultural land

1.2. Portland-Limestone Cements

Limestone cement is one of the most important types of cement that is used in the construction and finishing process and is widely used in the world, where limestone is added to clinker in specific quantities to obtain cement with a compressive strength of 42.5 MPa at 28 days, the

limestone cement is used in Palestine in plastering and brick bonding works.

Limestone, or calcium carbonate (CaCO_3), is one of the substances that has been added to Portland and mixed cements as an ingredient. As a result, Portland-limestone cement, clinker that has been combined with limestone, is produced.

Based on the amount of limestone added to the pure clinker, Portland-limestone cements are divided into different categories. Portland-limestone cements with limestone content ranging from 5% to around 40% are made and utilized in a number of nations worldwide, with the cement type CEM II/A with 5-20% limestone being the most popular in Europe [3].

In the most recent iteration of the European Standard EN 197-1 (2011), the clinker used to make PLC is the same clinker used to make normal Portland cement. The 27 prevalent varieties of cement that the standard recognizes are all permitted to have up to 5% of minor extra ingredients; the most common of these are limestone and cement raw meal.

In addition, four of the designated 27 types are Portland-limestone cements that permit higher proportions of limestone in two replacement level ranges, namely CEM II/A-L and CEM II/A-LL with 6 to 20% limestone and CEM II/B-L and CEM II/B-LL with 21 to 35% limestone in addition to the 5% [6].

According to EN 197-1 the Limestone use in cement production must contain at least 75% by weight calcium carbonate while the clay content, determined by the methylene blue test in accordance with EN 933-9, must not exceed 1.20 wt.%. The total organic carbon (TOC) content is measured in accordance with EN 13639:1999, it must not exceed 0.20 wt.% for LL type and 0.50 wt.% for L type [6].

Many countries have more stringent regulations regarding cement production because of the impact cement production has on the climate. For instance, the cement industry in the United States decided to standardize Portland-limestone cements within specifications of ASTM C595 [7].

1.3. Literature Review

Reusing stone waste in various industrial activities was discussed in Elham Khalilzadeh Shirazi's work, which offered several solutions for incorporating this industrial by product into building materials like cement, tiles, mortars, self-compact concrete (SCC), other cement-based goods, pavement, embankments, agglomerate marble, and the creation of glues and paints [8].

Nabil Al-Joulani's study, investigated how stone slurry waste from the stone-cutting business is used as a raw material for the manufacture of artificial stone. From the findings of this study, the following conclusions can be drawn:

1. The percentage of cement to stone powder, compaction pressure, and curing time all have a

significant impact on the compressive strength of artificial stones.

- By increasing the compaction pressure and cement percentage, absorption was reduced [9].

Digvijay S. Chouhan, et al. [10] studied the use of granite slurry waste as a variety of replacements for fine aggregates and cement in concrete has been examined in this paper. The research found that replacing fine aggregates with granite slurry waste results in a cohesive mix and decreases the workability of granite slurry concrete. When the replacement level exceeded 20%, poor workability was observed.

Nuno Almeida, et al. [11] suggested a number of technical solutions for incorporating this industrial by-product into building materials like cement, red ceramic bricks and tiles, resin-based materials, mortars and concrete, agglomerate marble, etc. or in other construction solutions like paving, embankments, and road construction. The nature of this waste also allows for additional applications in a number of industries, including but not limited to the paper industry, the ceramics industry (faience), agriculture, water treatment, and landfill sealing. The nature of this waste is mostly influenced by its origin in terms of minerals.

The main objectives of this work are to produce inexpensive, environmentally friendly Portland-limestone cement (PLC) specimens using the slurry of cutting stone industry with different percentages (5-15 wt.%), and then to investigate the effect of slurry cutting stone content on the properties of the produced cement and specimens.

2. Materials and Methods

2.1. Materials

Samples of cutting stones slurry were collected from five quarries in Palestine, while the clinker was supplied from

Jordan (Northern Cement company).

2.2. Methods

- The collected cutting stones slurry samples were dried in the oven dry at 115°C for 15 minutes to remove their moisture contents.
- The dried samples were tested chemically at (Birzeit University Laboratory, Palestine) to measure their chemical compositions.
- The fineness test on the chosen powder was carried out by using (Blaine Air permeability method according to EN 196-6), the test showed that powder has a fineness of 5000-6200 cm²/g, which means that no further crushing needs to be applied on the limestone powder.
- Sample of clinker which was grounded separately in (Sealing Grinding Machine GJ100-3) up to 4000 cm²/g fineness.
- Limestone powder, clinker and gypsum were mixed in different proportions, the percentages of each component in the prepared mixtures are shown in Table 1.

Table 1. The percentages of each component in the mixture

Item	Limestone powder	Clinker	Gypsum
Mix No.1	0%	95%	5%
Mix No.2	5%	90%	5%
Mix No.3	10%	85%	5%
Mix No.4	15%	80%	5%

- Compressive strength, soundness, shrinkage, consistency and setting time test were performed according to the European Specifications and flow for mortar according to ATM specification. The number of samples for each test is shown in the text below.
- Requirements of Portland limestone cement used in Palestine are showed in Table 2.

Table 2. Physical and mechanical properties of PLC cement used in Palestine according to EN 197-1

Portland limestone cement types	Limestone Content %	Strength Class	Compressive strength (MPa)		Initial setting time (min)	Soundness (Expansion) mm	CaCO ₃ % In Limestone
			2 days	28 days			
CEM II/A-L	6 to 20	42.5 N	≥20	≥42.5 ≤62.5	≥60	≤10	75≤
CEM II/B-L	21 to 35						

3. Results and Discussion

3.1. Chemical Analysis

5 samples from different queries located in west bank were chemically analyzed by ICP Method in order to investigate their chemical composition.

The preparation of limestone samples for elemental analysis is done according to the following steps:

1. Mix the sample thoroughly and draw a representative sample for analysis.
2. The sample is dried in an oven at 70°C.
3. The sample is sieved using 60-80 mesh sieve.
4. Take around 1 gram of the sieved samples (record the weight up to 4 digits (x.xxxx gm))
5. Ignite the weighed samples at 550-600 °C for 4.5 - 5 hours then cool at desiccator to room temperature.
6. Digest ash content in the same crucible directly with concentrated nitric acid and hydrochloric acid until solution is clear.
7. The digested sample is transferred into 50 ml volumetric flask and diluted with MQ water to the required volume.
8. Filter around 10 ml from the diluted solution using 0.45 Micrometer disk filter.
9. The sample runs on ICP against certified multi element standard

The chemical composition of each samples is shown in Table 3.

It can be seen from Table 3 that calcium oxide is the main constituent of the collected limestone samples, its content varies from 47.61 wt.% to 53.71 wt.%. The European specifications for using limestone in cement production require a minimum calcium carbonate content of about 75 wt.% [6]. The following equation can be used to calculate the calcium carbonate content of the collected limestone samples.

$$CaCO_3 \% = \frac{CaO\% * MW_{CaCO_3}}{MW_{CaO}} \quad (1)$$

Where the molecular weight of calcium carbonate and calcium oxide equals to 100 g/mole and 56 g/mole, respectively.

The calcium carbonate content of the collected samples is shown in Table 4.

Tables 3 and 4 show that the magnesium oxide and calcium carbonate content of all collected samples satisfy the European specifications. Table 3 also shows that the chlorides content is very low which is a very important indicator because chlorides are the most harmful compounds to both concrete and reinforcing steel. The chemical test shows that the collected limestone samples contain an acceptable value of calcium oxide, magnesium oxide and chlorides and so they can be used in this study.

Table 3. Chemical analysis of the limestone powder collected from different quarries in Weast Bank

Test results	Content (wt.%)				
	Quarry 1	Quarry 2	Quarry 3	Quarry 4	Quarry 5
CaO	47.61%	51.81%	53.71%	50.29%	48.52%
MgO	5.272%	2.445%	0.405%	2.457%	5.516%
Fe ₂ O ₃	0.223%	0.149%	0,035%	0.304%	0.136%
K ₂ O	0.033%	0.029%	0,015%	0.045%	0.04%
Al ₂ O ₃	0.196%	0.098%	0,034%	0.162%	0.099%
TiO	0.004%	0.001%	0.001%	0.003%	0.002%
Mn ₂ O ₃	0.005%	0.004%	0.003%	0.007%	0.005%
P ₂ O ₅	0.021%	0.015%	0.017%	0.020%	0.038%
Na ₂ O	0.030%	0.028%	0.030%	0.034%	0.037%
SiO ₂	2.370%	1.559%	0,424%	3.592%	1.238%
SO ₃	0.025%	0.045%	0.050%	0.017%	0.017%
Cl	0.048%	0.038%	0.792%	0.025%	0.030%
Fluorides	Not Detected				

Table 4. Calcium oxide and calcium carbonate content of the collected limestone samples

	Quarry 1	Quarry 2	Quarry 3	Quarry 4	Quarry 5
CaO%	47.61%	51.81%	53.71%	50.29%	48.52%
CaCO ₃ %	85.02%	92.52%	95.91%	89.80%	86.64%
Requirements EN-197-1 CaCO ₃ %	75≤				

3.2. Fineness Test Results

The fineness test was performed according to BS EN 196-6 [12]. The results showed that the powder was crushed to the required fineness (size), since cement fineness is usually in the range of (3000-5000 μm) [4]. The fineness of the powder was found to be in the range of (5500-6000 μm) which is a positive result from the economical point of view, because the powder does not need extra crushing which minimizes extra added cost. Also, it has been found that when limestone is ground into a powder with a surface area greater than ($> 500 \text{ m}^2/\text{kg}$ Blaine), it can be combined with portland cement to improve the cementitious system's particle size distribution, resulting in a decreased need for water and improved cement properties [13].

3.3. Compressive Strength of Prepared Samples

3 tests were conducted for each mix and the mixing ratio was as follows: 1350 grams of standard sand, 450 grams of cement and limestone powder according to the proportions shown in Table 1, and 225 grams of water according to the requirements of European Standard 196-3 [14]. Figure 2

shows that the addition of the limestone powder with 5 wt.% and 10 wt.% had no significant effect on the compressive strength of the cement, the compressive strength of cement decreased after 28 days from 55.3 to 55.2 and 54.5 MPa upon the addition of 5 wt. % and 10 wt.% limestone powder, respectively. While the addition of 15 wt.% lime stone powder decreased the compressive strength to 46.7 MPa, the addition of limestone affects the compressive strength because the limestone powder size is less than the cement particles, so when the limestone particles exist plentifully they may accumulate together and form stress concentration zones that can accelerate the fracture of the tested samples. Also the fillers can enhance the hydration of Portland cement by acting as nucleation sites [15].

The results obtained are consistent with those of other studies, and they can be interpreted as follows: the use of limestone increased the porosity of the paste fraction of mortars [13]; however, finer ground limestone could be combined with relatively coarse clinker to reduce the overall porosity in cement; this resulted in the densification of the microstructure and interfacial transition zone in systems where limestone was used, which positively affects strength [16,17].

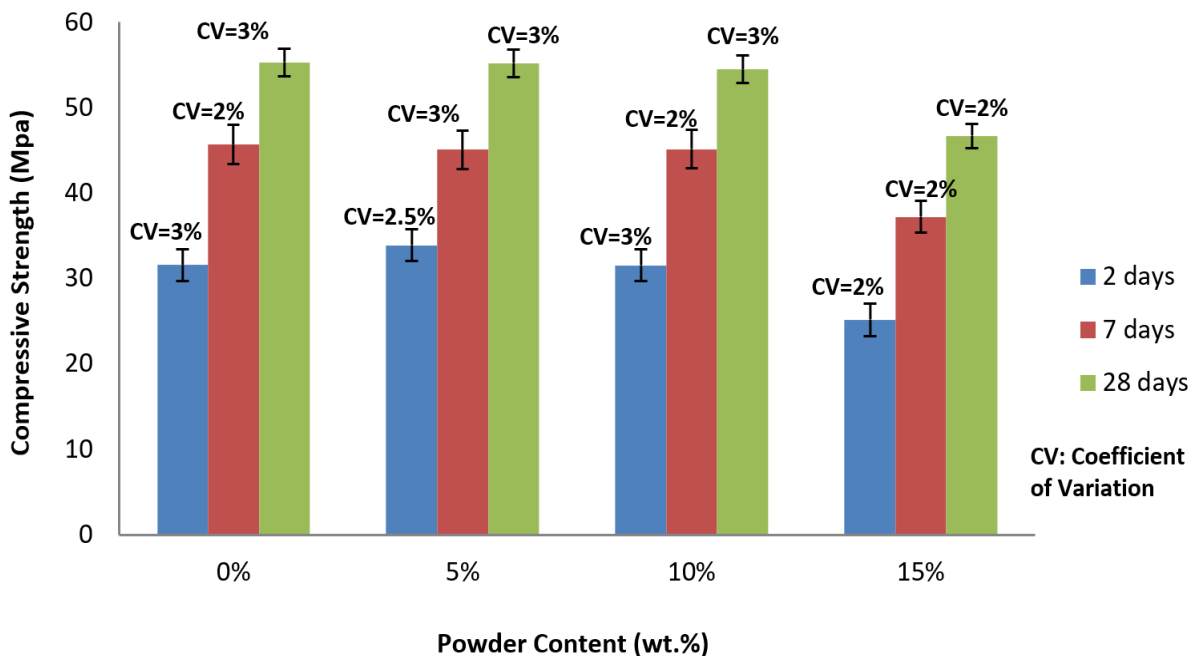


Figure 2. Compressive strength of the tested samples

3.4. Consistency and Setting Times of Prepared Samples

To determine the percentage of water needed to the setting time test, a consistency test was first performed, as this test expresses the amount of water needed for the reaction to occur, as well as the water needed for mixing in practices, to guarantee statistical representation three test performed for each mix.

The setting time and consistency were performed according to BS EN – 196-3 [18] for prepared samples. Table 5 shows that the addition of limestone powders up to 15 wt.% results in a slightly decrease in the percentage of the consistency of the prepared samples, it decreased from 29.5% to 27.0%, which is a good advantage in cement and concrete mixing because the water cement ratio and strength are inversely proportional to each other.

Setting time test is a test used to estimate the hydration time of the cement paste and the point at which the strength begins to develop, the long period of hydration time is most likely to be avoided, also very short hydration time is not preferred because it generates more heat which can be harmful to the cement paste and to concrete in general.

Table 4 shows that both the initial and final setting times of the prepared samples decreased with the addition of limestone powder, the initial and final setting times decreased around 35 and 30 minutes, respectively, when 15 wt.% limestone powder was added which means that there was a rapid development of early strength with increasing

limestone powder percentages. The decrease in the initial and final setting times may be related to three reasons, firstly, the small size of the added limestone powder is less than the size of cement particles which enables it to enter between cement particles, this results in a decrease in the pore content in the paste, secondly, the limestone powder acts as a nucleating agent for hydration process, and finally, the limestone powder tends to attract the hardened compounds from clinker particle surfaces which allow exes water to penetrate clinker particles as previous experiments indicate.

3.5. Effect of Limestone Slurry in Workability of Mortar

To determine the effect of limestone slurry in workability of mortar sets, samples were prepared in accordance with ASTM C109, the mixing ratios and flow are shown in Table 6. Cone flow method used according to ASTM C1437 Standard with this test a truncated cone was filled with mortar samples and placed at the center of the flow table, the truncated cone was removed leaving the mortar sample which was dropped continuously for 25 times. The diameter of the mortar along the four lines was measured and scribed in the table top, and the flow is the resulting increase in average base diameter of the mortar mass, expressed as a percentage of the original base diameter. The flow indicates the workability of the sample.

Table 5. Consistency and Setting times of The Prepared Samples

Samples	Consistency Test		Setting time		
	Percentages of Consistency	CV %	Initial time	Final time	CV %
95% clinker, 5% Gypsum	29.5%	3.0	145 min	210 min	2.0
90% clinker, 5% powder, 5% Gypsum	29.0%	3.0	115 min	195 min	2.0
85% clinker, 10% powder, 5% Gypsum	28.0%	3.0	110 min	185 min	3.0
80% clinker, 15% powder, 5% Gypsum	27.0%	3.0	100 min	170 min	2.0

Note: CV-Coefficients of Variation

Table 6. Mortar mixture proportions, and flow (workability) results

Mix	Sand (g)	Water (g)	Prepared cement composition (g)			Flow %
			Clinker	Gypsum	Limestone slurry	
Mix with 0 % slurry	1237.5	218.25	427.50	22.50	0.0	106
Mix with 5 % slurry	1237.5	218.25	406.13	22.50	21.37	108
Mix with 10 % slurry	1237.5	218.25	384.75	22.50	42.75	111
Mix with 15% slurry	1237.5	218.25	363.38	22.50	64.12	113

From the results above, we notice that the flow increases with the increase of the percentage of limestone powder, the reason for this can be explained by the fineness of the limestone powder, as the workability increases with increasing fineness.

3.6. Density of Cement with Limestone Slurry

To study the effect of limestone slurry on the density of cement, we conducted the test according to the ASTM C 188[19]. The test was conducted for three samples for each mixing ratio, as shown in Table 7.

The results of the density show that by increasing the

percentage of powder, the density increases. This can be explained by the fineness of the fine powder grains that fill the voids in the cement, which resulted in the densification of the microstructure of cement limestone.

3.7. Shrinkage Test Results

To study the effect of cement on shrinkage which is considered one of the important properties of cement used in plastering and finishing materials, 3 specimens were prepared for each mixing ratio according to proportions shown in Table 8. Figure 3 shows the percentage of shrinkage results of the prepared samples [20].

Table 7. Density test results of cement with different percentage of limestone powder

Mix No.1 without Slurry	Mix No.2 with 5 % Slurry	Mix No.3 with 10 % Slurry	Mix No.4 with 15 % Slurry
3.11(g/cm ³)	3.14(g/cm ³)	3.16(g/cm ³)	3.18(g/cm ³)
3.09(g/cm ³)	3.13(g/cm ³)	3.16(g/cm ³)	3.17(g/cm ³)
3.09(g/cm ³)	3.14(g/cm ³)	3.16(g/cm ³)	3.18(g/cm ³)
Average 3.103(g/cm ³)	Average 3.136(g/cm ³)	Average 3.16(g/cm ³)	Average 3.176(g/cm ³)

Table 8. Mixing proportions for shrinkage test

Samples	Mixing water W/C	Aggregate sand (g)	Weigh of cement and limestone (g)
95% clinker, 5% Gypsum	0.7	1350	450
90% clinker, 5% powder, 5% Gypsum	0.7	1350	450
85% clinker, 10% powder, 5% Gypsum	0.7	1350	450
80% clinker, 15% powder, 5% Gypsum	0.7	1350	450

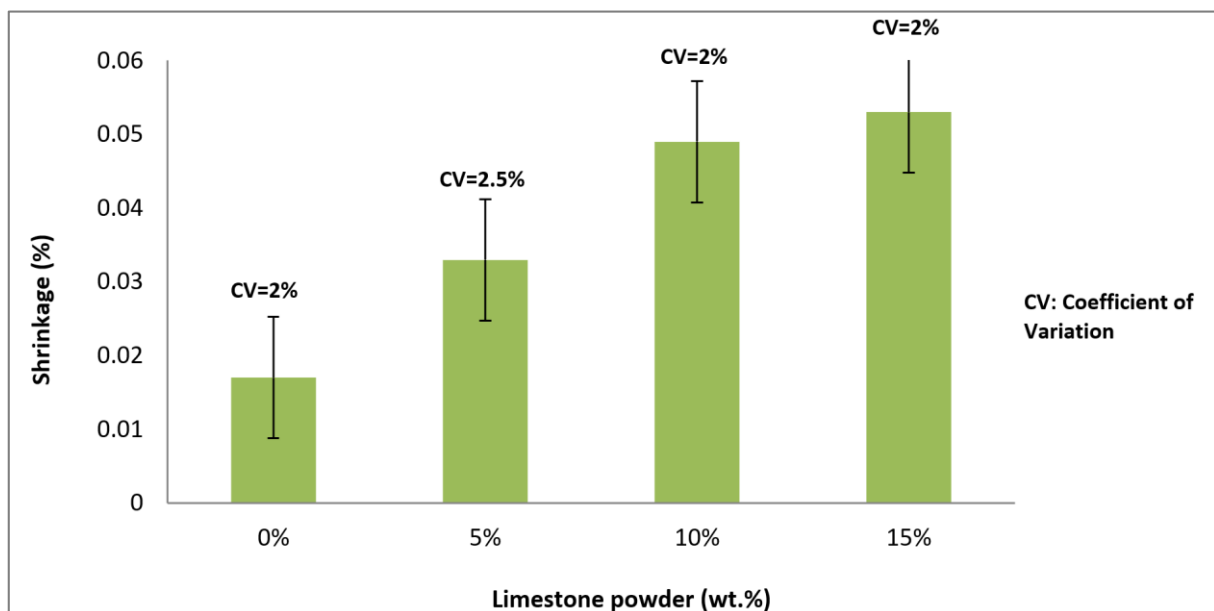


Figure 3. Shrinkage Test Results

It can be seen from Figure 3 that the loss of volume of the cement paste increased with the addition of limestone powder, which is the main cause of cracking that occurs after placing. As expected, the paste with higher percentages of fine materials tends to shrink more, since the limestone powder has less particle size and more surface area the powder absorbed the water in a quick manner from which that for the reference sample with 0% limestone. In fact, there are no standards limits for shrinkage according to BS EN 12390-16 but in comparison with 0 wt.% limestone powder, the samples containing limestone powder do not have very higher values.

3.8. Soundness Test Results

The soundness test measures the expansion of cement paste at extreme conditions which is considered as a vital sign of durability of cement and concrete, high expansion values in concrete are followed by cracking and deterioration of the construction elements. Furthermore, cracking improves the permeability of the harmful compounds like water and chlorides to the reinforcing steel in the construction elements. The data analysis of the soundness test exhibited promising results about using limestone powder in cement production, the addition of limestone powder up to 15 wt.% did not affect the expansion value of the cement samples, it had a value of 1.0 mm for all tested samples performed according to BS EN -196-3 [18], which is an acceptable value according to EN197 -1 because soundness becomes harmful when the expansion value exceeds 10 mm. The expansion value of the prepared samples remains constant due to the small size of limestone powder, which is smaller than the cement particles, so it was integrated with cement particles and filled the empty space between them rather than taking extra size.

4. Conclusions

The dried stone cutting industry slurry is utilizable in cement production industry and needs no further treatment and crushing. The chemical test of the dried slurry showed that it satisfied the EN197-1 requirements and also exhibited a promising result about cement durability due to the absence of harmful compounds which may restrict its use and negatively affect its durability. The utilization of stone cutting wastes in cement production significantly decreases its effects on the ecosystem components. The compressive strength of the blended cement decreased slightly when 5 wt.% and 10 wt.% of limestone powder were added while the decrease in the compressive strength was 15.5% when 15 wt.% of limestone powder was added. Increasing the limestone content increased the shrinkage in the cement paste but with small values which do not affect the concrete durability. The expansion in the cement paste remained the same until even at 15 wt.% limestone powder.

The utilization of stone cutting slurry in cement production is very profitable if applied in a large scale. The blended cement could be used in higher percentages of limestone powder at other nonstructural construction concrete. The use of stone cutting industry powder will reduce the carbon dioxide emissions by 10% if applied.

Based on the results, the cement produced with limestone slurry can be used in the production of many construction materials such as concrete bricks, mortar for bricks bonding, plaster and other finishing materials.

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