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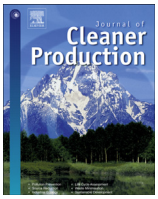

# Investigating chemical, physical and mechanical properties of eco-cement produced using dry sewage sludge and traditional raw materials

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## ABSTRACT

This work was performed to investigate the effect of using dry municipal sewage sludge as one of the raw materials in Portland cement production on the mechanical and durability properties of an environmentally sustainable cement (eco-cement). The specimens were prepared as one control and four different replacement level of dry sewage sludge as partial substitute of traditional raw materials (5.0, 7.5, 10.0 and 15.0 percent). Several physical, chemical and mechanical characteristics of eco-cement produced in this study were determined experimentally. The results indicated that chemical composition of all specimens were very similar to each other. Moreover, the specific gravity and fineness of specimens containing sludge were found to be lower and higher than control specimens, respectively. Initial and final setting times of eco-cement increased with the increase in replacement level of dry sewage sludge. In addition, with increasing the replacement level, the normal consistency, fluidity and the volume expansion values of specimens increased. It was also observed that for all eco-cement specimens, the compressive and flexural strengths were comparable to control specimens.

## 1. Introduction

Municipal sewage sludge, an inevitable side product of wastewater treatment process, is one of the wastes generated as a result of human activity in urban environments. The direct disposal of sludge from wastewater treatment plants can cause acute environmental problems due to existence of high amounts of pathogens, heavy metals and unstable organic matter in its composition. The per capita amount of solids sludge produced was reported to be 35–85 g per day (Davis, 1996). The presence of nutrients such as nitrogen and phosphorus in sewage sludge can justify the use of this substance as a biological fertilizer in agricultures. However, environmental issues and existence of strict standards and regulations have limited and complicated its usage (Rulkens, 2008; Świerczek et al., 2018). Since the sludge is dominantly composed of organic matters, it could be used as energy source as well. Presence of mineral elements such as silicon, aluminum, and iron in the chemical composition of municipal sewage sludge, makes it possible to use this material in cement production (Świerczek et al., 2018).

To date, some researches have been conducted regarding the effect of sewage sludge utilization on the environmental issues. These studies showed that using sewage sludge as a fuel in cement production processing in addition to providing dependable and low-cost source of energy, is advantageous to reduce the pollutant emissions and does not aggravate further health risks for inhabitants living near a plant. (Atienza-Martínez et al., 2018; Cieślik et al., 2015; Gálvez et al., 2007; Husillos Rodríguez et al., 2013; Nadal et al., 2009; Rovira et al., 2011; Zabaniotou and Theofilou, 2008).

Husillos Rodríguez et al. (2013) reported that using this material as replacement of raw materials in cement production (14%) could reduce consumption of silica sand, clay, limestone and fuel considerably. Lin et al. (2012) found that increasing the replacement level of dry sludge from 0.5 to 15.0%, helps the formation of $C_2S$ (2CaO.SiO2) and subsequently prolong the cement setting time. Combination of sludge with lime to produce cement like materials

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carried out by Tay and Show (1991). They concluded that the cement made from sludge satisfied the limiting values of standards and it could be used for common masonry operations.

Furthermore, several investigations were carried out by researchers to incorporate sewage sludge ash with cementitious materials. In this regard, Lin and Lin (2005), used a combination of three types of waste materials, including municipal wastewater sludge, water purification sludge, and steel industry slag as alternatives of raw materials for cement production. In general, they concluded that sludge ash and ferrate waste could be used to replace up to 20% of raw mineral in clinker production process. In 2016, Chakraborty et al. (2017) combined sewage sludge ash with quicklime and blast furnace slag as a cementitious materials to produce mortars. Their results showed that this combination improved the physical and mechanical properties of the produced mortar. Some investigations have also focused on environmental impacts of sewage sludge ash. It was deduced that disposal of these hazardous materials in landfills has damaging effects on the environment (Donatello et al., 2010; Lapa et al., 2007; Li et al., 2018). There are also some studies dealing with the physical and mechanical properties of cementitious materials produced using sewage sludge ash (Chen and Poon, 2017; Garcés et al., 2008; Naamane et al., 2016; Piasta and Lukawska, 2016). These studies indicated that although the use of sewage sludge ash has some negative effects on the physical and mechanical characteristics of cement, it is economically feasible to utilize this materials as a substitution of a portion of raw materials for different applications.

The purpose of this study was to investigate different characteristics of cement produced by combination of dry municipal sewage sludge and traditional raw materials. Physical, chemical and durability tests were performed on specimens prepared with different replacement levels of dry sewage sludge and mechanical properties of the specimens were performed after 3, 7 and 28 days of curing.

## 2. Materials and methods

### 2.1. Materials

#### 2.1.1. Sewage sludge

For production of eco-cement (Portland cement containing dry sewage sludge), sludge was collected from the sewage treatment plant in Bojnurd (a city located in north east of Iran). The waste water treatment system in this plant is extended aeration; a version of activated sludge system. The wet sludge was collected from drying beds, and dried at 105 ± 5 °C in laboratory. Afterwards, the dried sludge was kept in a closed bag. To produce eco-cement, dry sludge was then powdered by a mill and used as partial substitute of traditional raw materials.

#### 2.1.2. Raw materials

The clinker production materials in this study, included limestone, marl, bauxite and iron ore, which were taken from quarries around the city of Mashhad (a city located in north east of Iran) and used in Mashhad Cement Company. In the first step, all raw materials were crushed by stone crusher and dried in an oven at 105 ± 5 °C for 24 h. Then, they were milled until all the particles were passed through a #100 sieve (particles smaller than 150 μm). Then, the dried samples of the materials were wetted with a suitable liquid (especially industrial methylated spirits to avoid migration of water-soluble components), in evaporating basins, until the particles agglomerate significantly. After that, small subsamples were worked by hand to form balls of about 8 mm diameter. The subsamples were then oven-dried in platinum boats at 105 °C. After drying, the boats were placed in the center of the pre-heated tube furnace. Typically the furnace would be pre-heated to 700 °C and different samples would be heated at a rate of 10 °C per minute to temperatures of 1400 °C, 1450 °C and 1500 °C and left at that temperature for 20 min. Then, the samples were removed from the furnace and cooled in a desiccator. The clinker produced was prepared for chemical analysis. The chemical composition of raw materials which were conventionally used for the production of the clinker, was obtained using X-ray fluorescence spectroscopy (XRF) and inductively coupled plasma atomic emission spectroscopy (ICP-AES) test methods and reported in Table 1. Also, the LOI (loss on ignition) test which was conducted on these materials, is explained in the following.

Furthermore, grading and physical properties of the sand used for producing the cement pastes are listed in Table 2 and Table 3, respectively.

In order to calculate the combination ratio of clinker production raw materials, a simple method, which has been previously suggested by other researchers was used (Lin and Lin, 2005; Lin et al., 2012; Yen et al., 2011; Zhou et al., 2012). In this method, several ratios such as silica modulus (SM), iron modulus (IM), and lime saturation factor (LSF) need to be calculated. Calculation of the ratios is performed using the following equations and based on the chemical composition of the raw materials:

$$SM = SiO_2/(Al_2O_3+Fe_2O_3),\ 2.25 < SM < 2.35 \tag{1}$$

$$IM = (Al_2O_3/Fe_2O_3),\ 1.3 < IM < 1.7 \tag{2}$$

$$LSF = CaO/(2.8SiO_2 + 1.18Al_2O_3 + 0.65Fe_2O_3),\ 0.9 < LSF < 0.96 \tag{3}$$

#### 2.1.3. Eco-cement clinker

In this work, five different mixing designs containing different dry sludge replacement levels were considered to investigate the effect of dry sludge as raw material on physical, chemical and mechanical properties of eco-cement. The specimens are named as follows:

PC: ordinary Portland cement without dry sewage sludge (as control)
ECX: eco-cement produced by replacing X% of cement raw materials with dry sludge.

For example, in the EC10, 90 g of traditional clinker production materials and 10 g of dry sewage sludge were used for production of 100 g of eco-cement clinker.

It should be noted that in cement production process, to control the setting time of produced cement, 5% gypsum was added to all eco-cement clinker specimens, and ground by a disc mill for 15 min. Blaine fineness of all specimens were between 4700 and 5200 cm$^2$/g.

### 2.2. Specimen preparation

The mix design of standard mortar for preparation of compressive and flexural strength test specimens was determined according to ASTM C 348. The composition (by weight) of mortar component consists of one part of cement, 2.75 part of standard sand and 0.485 part of water (the water to cement ratio is 0.485).

Specimens were also prepared and cured in accordance with ASTM C 192. The produced specimens were wrapped in a damp cloth for 24 h, and then demolded and immersed in the saturated lime water at the temperature of 28 ± 2 °C.

**Table 1**
Major compositions of clinker raw materials in terms of oxides.

| Chemical composition | Limestone | Marl | Bauxite | Iron ore | Sewage sludge |
|---|---|---|---|---|---|
| $SiO_2$ (%) | 2.56 | 26.40 | 11.28 | 27.00 | 9.30 |
| $Al_2O_3$ (%) | 0.16 | 4.70 | 43.66 | 6.50 | 2.53 |
| $Fe_2O_3$ (%) | 0.14 | 0.36 | 21.74 | 53.00 | 1.43 |
| CaO (%) | 51.23 | 31.30 | 6.75 | 4.50 | 12.06 |
| MgO (%) | 1.73 | 2.62 | 0.99 | 0.40 | 0.91 |
| $K_2O$ (%) | 0.11 | 0.60 | 0.08 | 0.10 | 0.39 |
| $Na_2O$ (%) | 0.37 | 0.45 | 2.50 | 0.22 | 0.62 |
| $SO_3$ (%) | 0.28 | 1.67 | 0.01< | 0.18 | 1.07 |
| $P_2O_5$ (%) | 1.20 | 1.09 | 0.25 | 0.55 | 0.22 |
| $TiO_2$ (%) | 0.86 | 0.28 | 0.67 | 0.52 | 0.11 |
| MnO (%) | 0.12 | 0.05 | 0.11 | 0.08 | 0.01< |
| Cl (%) | 0.01< | 0.01< | 0.01< | 0.01< | 0.01< |
| LOI (%) | 40.13 | 29.62 | 11.09 | 6.89 | 71.22 |
| As (mg/kg) | 0.25 | 0.04 | 0.09 | 0.18 | 0.05 |
| Ba (mg/kg) | 14.03 | 9.01 | 19.54 | 20.2 | 5.15 |
| Co (mg/kg) | [a] | 2.64 | 3.42 | 35.21 | [a] |
| Cr (mg/kg) | [a] | 15.63 | 24.95 | 26.71 | [a] |
| Cu (mg/kg) | [a] | [a] | 2.76 | [a] | 0.76 |
| Ni (mg/kg) | 0.06 | 6.54 | [a] | 10.51 | 1.64 |
| Pb (mg/kg) | 8.56 | 4.27 | 7.95 | 26.74 | 0.73 |
| Sb (mg/kg) | [a] | 1.26 | 5.45 | 12.65 | [a] |
| Sr (mg/kg) | 134.65 | 265.03 | 247.89 | 354.65 | 5.85 |
| Zn (mg/kg) | 32.65 | 87.96 | 45.67 | 168.93 | 1.95 |

[a] Not detected.

**Table 2**
Standard sand gradation.

| Sieve size (mm) | Weight retained on (%) |
|---|---|
| 2.00 | 0 |
| 1.60 | 7 ± 5 |
| 1.00 | 33 ± 5 |
| 0.50 | 67 ± 5 |
| 0.16 | 87 ± 5 |
| 0.08 | 99 ± 1 |

**Table 3**
Physical properties of the standard sand.

| $\gamma$ ($kg/m^3$) | $G_{FA}$ | $W_{FA}$ (%) | $W_{SSD,FA}$ (%) | FM |
|---|---|---|---|---|
| 1706 | 2.64 | 0.2 | 0.9 | 2.86 |

where $\gamma$, $G_{FA}$, $W_{FA}$, $W_{SSD,FA}$ and FM are specific dry weight, dry density, moisture content, water absorption and fineness modulus of the sand, respectively.

### 2.3. Test methods

The experiments were performed on both sludge and eco-cement pastes, which were produced from dry sewage sludge.

- Sewage sludge moisture content: To determine the moisture content of the sludge, a sample of wet sludge was poured into a crucible and weighed. Then, it was placed inside an oven with a temperature range of 103–105 °C, until a relatively constant weight was achieved (variation in weight was less than 3%) (Apha, 1998). The loss of weight (difference between the initial and the final weights of the samples) was used to calculate the moisture content.
- Fixed and volatile solid materials: To determine the fixed and volatile solids of sewage sludge, a sample of dry sludge was poured into a crucible and weighed. Then, it was placed inside a kiln with a temperature of 550 °C, until a relatively constant weight was achieved (variation in weight was less than 3%) (Apha, 1998). The loss of weight (difference between the initial and the final weight of the samples) was used to calculate the volatile solid materials. The remaining material in the container indicated the fixed solids. The determination is beneficial in control of wastewater treatment plant operation because it approximately offers the amount of organic matter present in the sledges. Although this method cannot precisely discern between organic and inorganic matters because in some cases the loss in weight can be due to decomposition or volatilization of some mineral salts.
- Chemical composition: Approximate percentage of the oxides of calcium, silicon, iron, aluminum, magnesium and sulfur, existing in the cement was evaluated by means of XRF test. In addition to XRF analysis, the wet chemistry method was also used for determination of the cement chemical analysis. Based on wet chemistry analyses method, the amount of the four main cement components including $C_3S$ ($3CaO.SiO_2$), $C_2S$ ($2CaO.SiO_2$), $C_3A$ ($3CaO.Al_2O_3$) and $C_4AF$ ($4CaO.Al_2O_3.Fe_2O_3$) were determined using the results of oxides compositions and Bogue equations which have been given in ASTM C150.
- Loss on ignition (LOI): LOI was determined by heating a certain amount of cementitious material in temperature range of 900–1000 °C until its mass remained unchanged. Then, the mass loss was measured. The ASTM C114 method was used to determine the LOI.
- Cement density: Cement density was measured according to ASTM C188. Density does not indicate the quality of cementitious materials and it is only used to determine the absolute volume of cement particles which used in mix design.
- Fineness: Fineness of cement particles affects the hydration reaction rate and its released heat. The finer the materials, the higher hydration rate and consequently the higher strength growth rate that the cementitious materials are supposed to possess. The effect of cement fineness on strength essentially appears within the first seven days (Waddell, 1974). In this study, the degree of fineness was measured according to ASTM C204, i.e. by measuring the residue on a 45 μm sieve and performing the Blaine air permeability test.
- Normal consistency of hydraulic cement: The aim of this test is to determine the amount of required water for preparation of a cement paste, which was used in other experiments as optimum

water to cement ratio. This test was carried out in accordance with ASTM C187.

- Flow test: Fluidity is the relative mobility of fresh mixture (cement paste or mortar) or its ability to flow. The test mortar was made in such a way that the value of water to cement ratio was constant or the flow rate was within a certain range. The consistency of the test mortar is determined using a flow table, according to ASTM C230.
- Setting time: Initial and final setting times indicate the normal or non-normal hydration reactions of the cement paste within the initial hours. Various factors such as temperature, cement fineness, additive materials, and the water to cement ratio, could affect the setting time of the cement paste. In this study, the setting time of Portland cement with and without dry sewage sludge was evaluated in accordance with ASTM C191.
- Compressive strength: It is the most important mechanical property of hardened concrete and cement paste in terms of design. For most cements, compressive strength is an important characteristic which indicates its quality, and the quality of concrete made from it. Compressive strength of cement specimens was measured according to BS 1881-116 standard, at curing ages of 3, 7 and 28 days.
- Flexural strength: This characteristic of cement paste is important for items such as unreinforced concrete and concrete pavement. In this study, flexural strength was evaluated at three curing ages (3, 7 and 28b days), in accordance with ASTM C78.
- Autoclave expansion of hydraulic cement: The cement health depends on volume stability of hardened cement paste after curing. When the raw materials that are charged into the kiln contain extra lime which cannot react with the mineral oxides and gypsum, it will remain in free lime state and cause the cement paste to expand after its hardening process. It is worth mentioning that the cement paste volume should not change after its curing and hardening, since it can trigger cracks and their growth inside the cement paste. In this study, Autoclave was used to determine the health of cements with and without dry sewage sludge, according to ASTM C 151.

## 3. Results and discussion

### 3.1. *Sewage sludge properties*

Moisture content of wet sewage sludge, and volatile and fixed solid contents of the dry sewage sludge are presented in Table 4. The fixed and volatile solid material test was performed on the dry sewage sludge which was obtained after moisture content test. Based on the results, the values of average moisture contents of wet sludge, volatile and fixed solid of dry sewage sludge used in this study were 33.7%, 69.7% and 30.3%, respectively. As previously mentioned, the volatile content approximately gives the organic matter of sludge which is useful in control of wastewater treatment plant.

### 3.2. *Clinker blend proportioning*

For production of the clinker containing dry sewage sludge, the replacement levels of dry sewage sludge were determined. Then, according to chemical analysis of dry sludge, other raw materials including lime, marl, bauxite and iron ore were selected so that the coefficients for IM, SM and LSF became 1.5, 2.3 and 0.92, respectively.

Table 5 presents the proportion of different raw materials for eco-cement clinker by weight. As it can be seen, replacing 15% of total raw materials by dry sludge in clinker production increased the required amount of Lime by 6% and reduced consumption of marl, bauxite, and iron by 35, 50, and 17%, respectively.

### 3.3. *Chemical compositions*

Chemical analysis of ordinary Portland cement and different eco-cements investigated in this study, which were determined using XRF and ICP-AES test methods are shown in Table 6. As the results of analyses show, the present values of main oxides components of these eco-cements (in all replacement ratios) are similar or very close to Portland cement oxide composition. However, it is observed that the $C_2S$ formation intensifies, as the dry sludge replacement level increases. This increase is insignificant up to 7.5% replacement level, which is in line with the results obtained by Lin et al. (2012).

As shown in Table 6, in general, the element contents (especially heavy metal contents such as Cr, Zn, Ni and Cu) of eco-cement specimens either decreased or not considerably changed with increasing replacement level of dry sewage sludge. Such a results can be acceptable according to the results presented in Table 1. Based on the data reported in Table 1, the elements content in sewage sludge is generally lower than other cement production raw materials (especially limestone and marl which constitute the main part of traditional raw materials). Thus, with increase in replacement level of sewage sludge, the values of elements content reduced in produced eco-cements. All in all, it can be expected that such insignificant changes of elements content in eco-cements compared to ordinary Portland cement not have great impact on the cement characteristics.

Also, using the amount of oxides given in Table 6 and considering the Bogue equations which obtained from ASTM C150, the percentages of four main components ($C_2S$, $C_3S$, $C_3A$, $C_4AF$) of ordinary cement and produced eco-cements were calculated and reported in this table. According to these results, it is apparent that the amount of $C_3S$ and $C_3A$ decreased, and the content of $C_2S$ and $C_4AF$ increased, with increase in sewage sludge replacement revel. The efficacy of these decreases and increases on properties of produced eco-cements such as setting time and mechanical strength are discussed in the following sections.

Furthermore, the results presented in Table 6 indicate that with increase in sludge replacement level, the amount of MgO increases,

**Table 5**
Weight percent of tested clinker components.

| Blending treatment code | Lime | Marl | Bauxite | Iron | Dry sewage sludge |
|---|---|---|---|---|---|
| PC | 48.5 | 48.70 | 1.00 | 1.8 | – |
| EC5 | 49.7 | 42.50 | 1.00 | 1.8 | 5 |
| EC7.5 | 50.5 | 39.65 | 0.85 | 1.5 | 7.5 |
| EC10 | 50.9 | 36.90 | 0.70 | 1.5 | 10 |
| EC15 | 51.3 | 31.70 | 0.50 | 1.5 | 15 |

**Table 4**
Moisture content of wet sewage sludge in 105 °C, and volatile and fixed solid contents of dry sludge in 550 °C.

| | Moisture content of wet sludge (%) | Volatile solid content of dry sludge (%) | Fixed solid content of dry sludge (%) |
|---|---|---|---|
| Average (3 samples) | 33.7 | 69.7 | 30.3 |
| Standard Deviation | 3.29 | 1.55 | 1.55 |

**Table 6**
The effect of replacing clinker production materials by dry sludge on chemical analysis of produced eco-cement (%).

| | PC | EC5 | EC7.5 | EC10 | EC15 |
|---|---|---|---|---|---|
| $SiO_2$ (%) | 21.48 | 21.50 | 21.52 | 21.56 | 21.61 |
| $Al_2O_3$ (%) | 4.72 | 4.69 | 4.60 | 4.59 | 4.50 |
| $Fe_2O_3$ (%) | 3.87 | 3.90 | 3.93 | 3.95 | 3.98 |
| CaO (%) | 62.73 | 62.70 | 62.50 | 62.41 | 62.35 |
| MgO (%) | 2.73 | 2.80 | 2.94 | 2.98 | 3.07 |
| $SO_3$ (%) | 2.40 | 2.39 | 2.40 | 2.41 | 2.42 |
| $Na_2O$ (%) | 0.38 | 0.41 | 0.47 | 0.56 | 0.68 |
| $K_2O$ (%) | 0.25 | 0.28 | 0.31 | 0.33 | 0.39 |
| $P_2O_5$ (%) | 1.13 | 1.11 | 1.12 | 1.09 | 1.08 |
| $TiO_2$ (%) | 0.57 | 0.57 | 0.58 | 0.58 | 0.58 |
| MnO (%) | 0.09 | 0.08 | 0.08 | 0.08 | 0.08 |
| Total alkalinity (%) | 0.54 | 0.59 | 0.67 | 0.77 | 0.93 |
| Cl (%) | 0.01 | 0.01 | 0.01 | 0.01 | 0.01 |
| As (mg/kg) | 34.12 | 33.02 | 32.71 | 32.04 | 30.89 |
| Ba (mg/kg) | 0.14 | 0.15 | 0.16 | 0.16 | 0.17 |
| Co (mg/kg) | 11.75 | 12.21 | 12.83 | 12.61 | 13.00 |
| Cr (mg/kg) | 1.95 | 1.79 | 1.60 | 1.53 | 1.38 |
| Cu (mg/kg) | 8.34 | 7.37 | 6.81 | 6.34 | 5.48 |
| Ni (mg/kg) | 0.03 | 0.15 | 0.27 | 0.27 | 0.39 |
| Pb (mg/kg) | 3.40 | 3.27 | 3.32 | 3.14 | 3.07 |
| Sb (mg/kg) | 6.79 | 6.75 | 6.73 | 6.63 | 6.55 |
| Sr (mg/kg) | 0.90 | 0.82 | 0.74 | 0.69 | 0.62 |
| Zn (mg/kg) | 201.38 | 189.39 | 182.44 | 175.32 | 162.54 |
| $C_3S$ ($3CaO.SiO_2$) | 48.04 | 47.95 | 47.52 | 46.86 | 46.77 |
| $C_2S$ ($2CaO.SiO_2$) | 25.34 | 25.46 | 25.84 | 26.46 | 26.67 |
| $C_3A$ ($3CaO.Al_2O_3$) | 5.96 | 5.82 | 5.54 | 5.48 | 5.19 |
| $C_4AF$ ($4CaO.Al_2O_3.Fe_2O_3$) | 11.77 | 11.86 | 11.95 | 12.01 | 12.11 |

as it could be anticipated considering high amount of MgO in sewage sludge. Nevertheless, even at the maximum replacement level of 15%, the amount of MgO in the cement was 3.07% and less than the maximum authorized amount in ASTM C150 standard (6%).

The result of Chemical analysis also showed that the average amount of sodium and potassium oxides and consequently, the total alkalinity ($Na_2O + 0.658K_2O$) of the eco-cement clinker, increases with increasing the dry sludge replacement level. Alkaline components are important part of cement composition due to their association with silica aggregates destructive chemical reactions. Hence, ASTM C 150 standard limits the total cement alkalinity to 0.6% for Portland cement type II. Altogether and based on the acquired results, it can be concluded that for replacement level of more than 5%, produced eco-cement does not meet the requirements of the maximum allowable alkalinity. Thus, if necessary, measures should be taken to reduce alkalinity or to avoid using silica containing aggregates.

### 3.4. *Loss on ignition (LOI)*

The values of LOI of eco-cement specimens with different sludge replacement levels is indicated in Fig. 1. Obtained results show that by increasing the replacement level of sludge, the LOI value of eco-cement increases. The LOI values of the eco-cement with the highest replacement level of 15% in this study was equal to 1.51% and less than the maximum acceptable value for Portland cement in ASTM C150 (3%).

### 3.5. *Specific gravity*

Fig. 2 depicts the specific gravities of the eco-cements produced in this research. As can be seen, higher levels of dry sludge replacement lead to decrease in cement specific gravity. This loss is probably due to increase in the porosity of cement particles caused by gas trapping during clinker production process, which stem from combustion of organic matters of sludge. The reduction in the specific gravity of cement containing dry sewage sludge compared to ordinary cement trigger a need for increase in the amount of water to achieve proper workability.

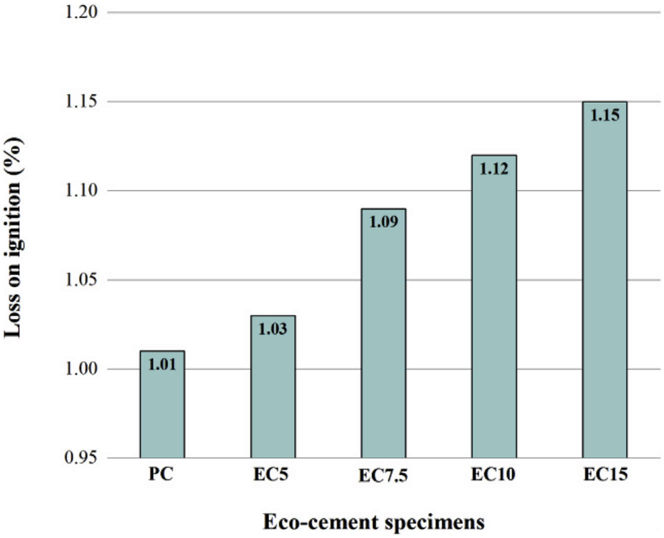

**Fig. 1.** The effect of replacing clinker production material by dry sludge on LOI.

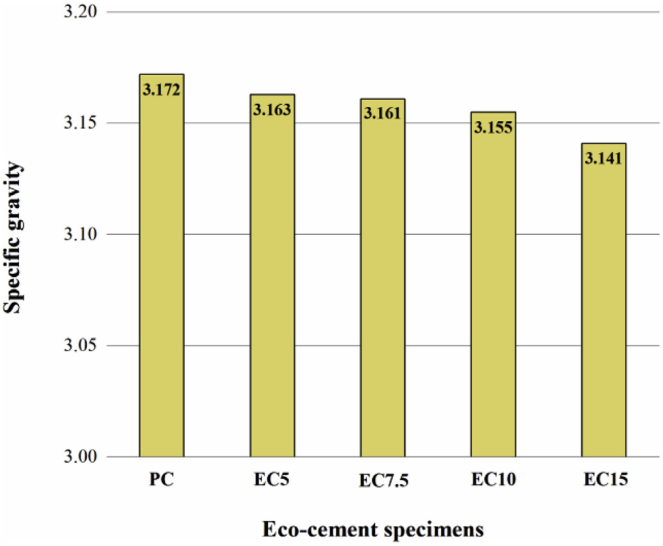

**Fig. 2.** The effect of replacing clinker production material by dry sludge on specific gravity.

### 3.6. *Free lime*

The amounts of free lime in cement specimens investigated in this study demonstrates in Fig. 3. These values were measured using modified Franke test method for determination free lime content in accordance with ASTM standard test method C114. The results show that the amount of free lime in cement pastes increases with increasing the dry sludge replacement level. In the highest replacement level of dry sludge (15%), the amount of free lime is 1.1% and within the acceptable range suggested by ASTM C150 (0.5–2.5%). Increment of the free lime content in the specimens containing higher amount of dry sewage sludge is mainly due to the fact that the dry sludge contains significant amount of CaO,

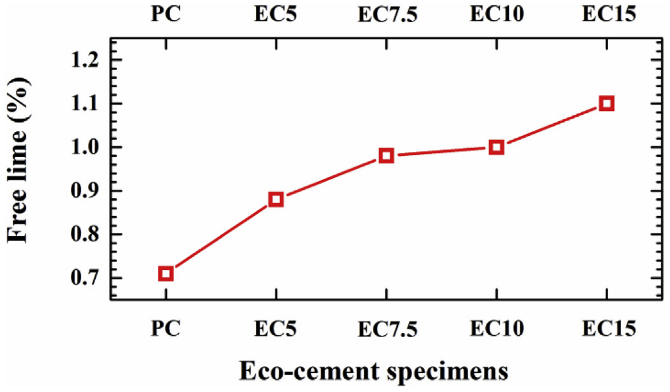

**Fig. 3.** The effect of replacing clinker material by dry sludge on free lime content.

so that the amount of free lime produced in clinker production is exceeded the required amount for reaction with $SiO_2$, $Fe_2O_3$ and $Al_2O_3$. Comparably, a study conducted by Zhou et al. (2012) on using the alkaline white mud as a part of raw materials for the production of cement, illustrated that increasing the content of such containing lime mud led to increase the free lime amount in the raw meal. Therefore, it is expectable that increasing the lime content used in mixtures containing dry sludge in this study, will increase the amount of free lime in the eco-cement paste, accordingly.

In addition, because of the heterogeneity of the raw meal mix, there may be extra CaO in some areas of cement matrix, while there is a scarcity of CaO in other areas (Waddell, 1974). In should be noted that, free lime can have a negative effect on cement behavior because it can give rise to expansion during hydration reaction as CaO converts to $Ca(OH)_2$ (Husillos Rodríguez et al., 2013).

### 3.7. Fineness

Two methods including residue on a 45-micron sieve and Blaine permeability were used to measure fineness of the produced cement in this study. The results are shown in Fig. 4. Based on these results, it can be observed that by increasing the dry sludge replacement level (keeping the milling time constant), the fineness of the specimens increases, using either methods. Increasing the fineness of eco-cement can accelerate hydration reactions and increase specific surface area of specimens. Also, more fineness leads to more workability of mortar due to the lubricate effect.

### 3.8. Normal consistency and fluidity

Cement paste with normal consistency is used to determine cement setting times and conduct cement health tests. Normal consistency is determined using inverted cone test (ASTM C187). Cement mortar with conventional fluidity is used to make specimens for compressive and flexural strengths tests. Conventional fluidity of a mortar is measured by flow table (ASTM C143). Therefore, for each combination of cement and dry sewage sludge, the amount of water needed to make a cement paste of normal consistency and cement mortar of conventional fluidity were determined as shown in Fig. 5.

Results indicated that by increasing the sludge replacement level, more water needed to achieve normal consistency and conventional fluidity. This outcome can be explained by the increase in the fineness value and porosity of eco-cements with higher sludge replacement levels as reported by other researchers as well (Garcés et al., 2008; Naamane et al., 2016; Tay and Show, 1991).

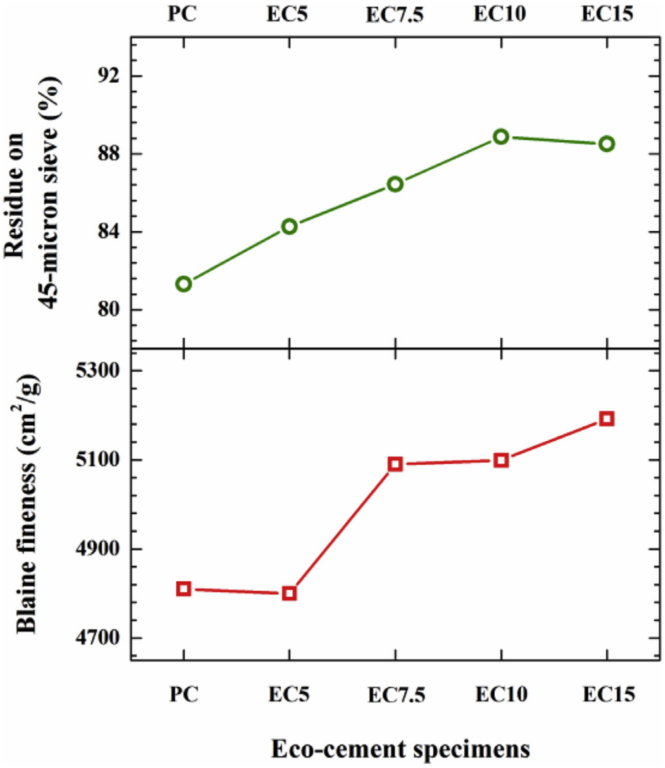

**Fig. 4.** Fineness of Portland cement and eco-cements.

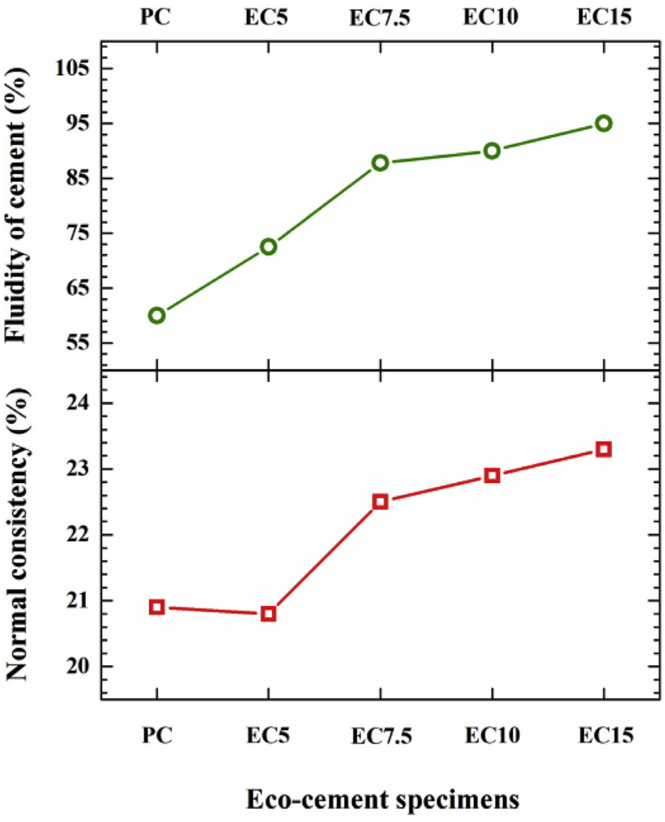

**Fig. 5.** Comparison of normal consistency and fluidity of Portland cement and eco-cements.

### 3.9. Setting times

The initial and final setting times for all eco-cement pastes in this study are depicted in Fig. 6. The results show that the initial and final setting times of Portland cement type II were 130 and 170 min, respectively. However, the setting times for all eco-cement pastes became longer compared with setting times of ordinary Portland cement paste as the replacement level increased. The previous investigators has also reported the same results (Lin et al., 2012).

The effect of increasing dry sludge replacement level on $C_2S$, $C_3A$ and $C_3S$ phase formation is also shown in Fig. 6. The results indicate that by increasing the replacement level of sewage sludge, the amount of $C_3S$ and $C_3A$ decreased, while the $C_2S$ content increased. The reducing $C_3S$ and increasing $C_2S$ can affect the setting time and strength development of produced eco-cement specimens. Other researchers similarly reached to these results (Lin et al., 2012; Naamane et al., 2016). It is important to note that the chemical reactions of $C_3A$ and $C_3S$ in the first few days of the hydration process of cement, generates excessive heat. Therefore, the decrease in the amount of $C_3A$ and $C_3S$ phases results in an increase in initial setting time. As can be seen in Fig. 6, with increase in the sludge replacement level, the $C_3A$ and $C_3S$ values decreased. This reduction of amount of $C_3S$ content in the produced eco-cement, lead to delay hydration process and prolongation of the initial setting times. Moreover, in another possible explanation, retarding effect on setting time as well as hydration reaction can attributed to the high amount of $C_2S$. These results are in line with those presented by other investigations (Lin et al., 2012).

Furthermore, prolongation of the initial and final setting times could be driven from the presence of some minor elements in dry sewage sludge, such as chromium, phosphorus and zinc, which can affect the hydration time (Cyr et al., 2007; Stephan et al., 1999).

### 3.10. Compressive and flexural strengths

Fig. 7 and Fig. 8 demonstrate the values of compressive and flexural strengths of all mortar specimens investigated in this study, respectively. Based on the presented results, it can be

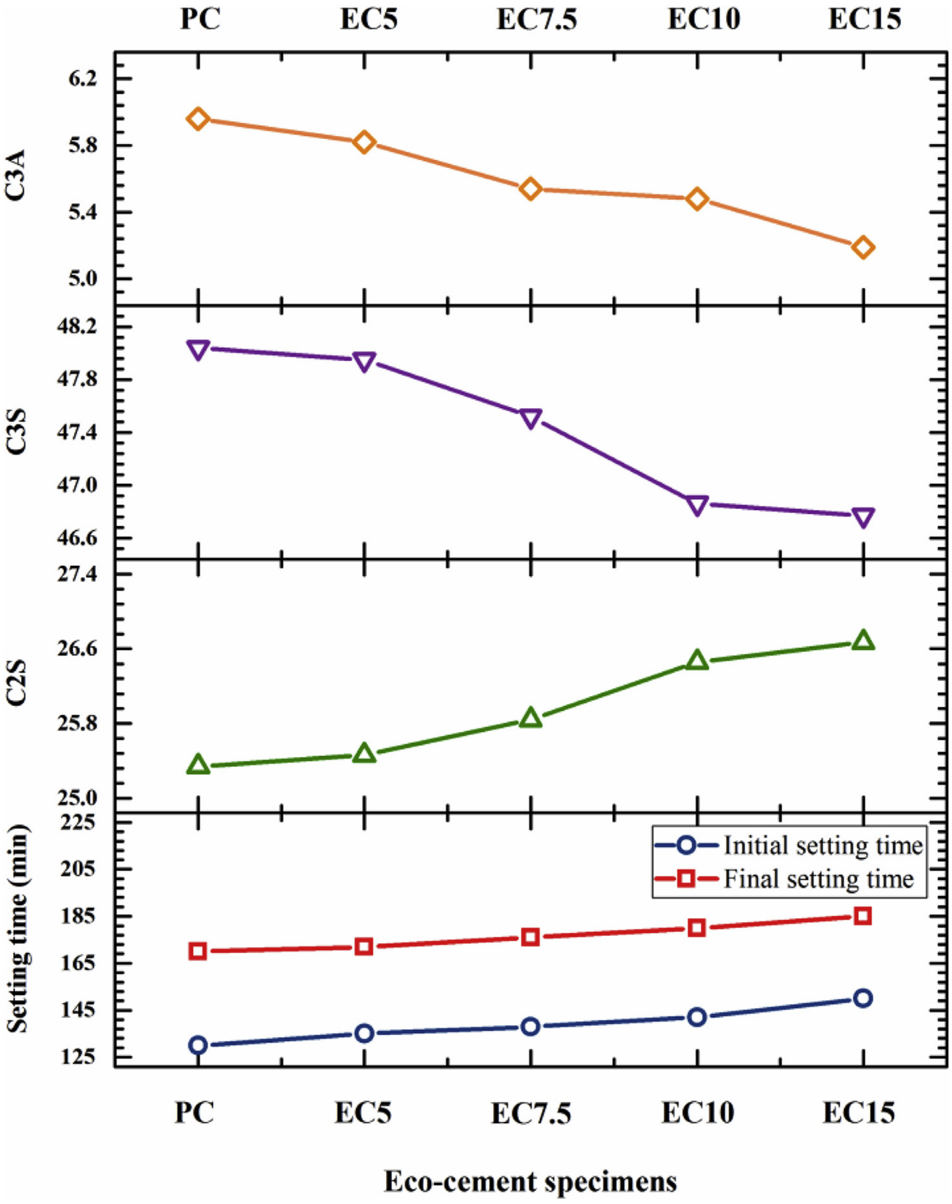

**Fig. 6.** The effect of different replacement levels of dry sewage sludge on initial and final setting times, $C_2S$, $C_3S$ and $C_3A$ contents of eco-cement specimens.

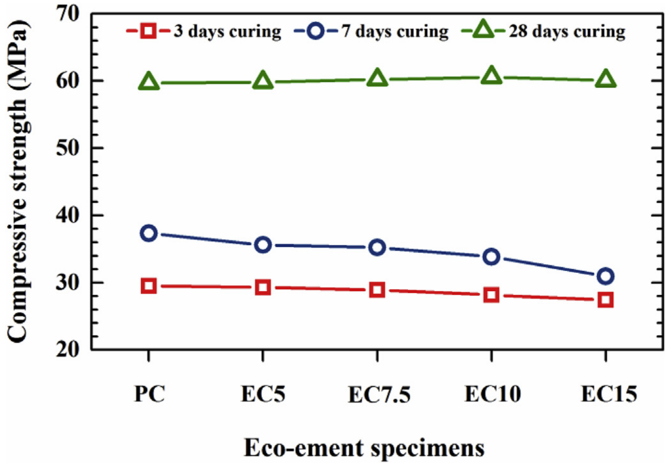

**Fig. 7.** The effect of different replacement levels of dry sewage sludge and curing periods on compressive strength of eco-cement specimens.

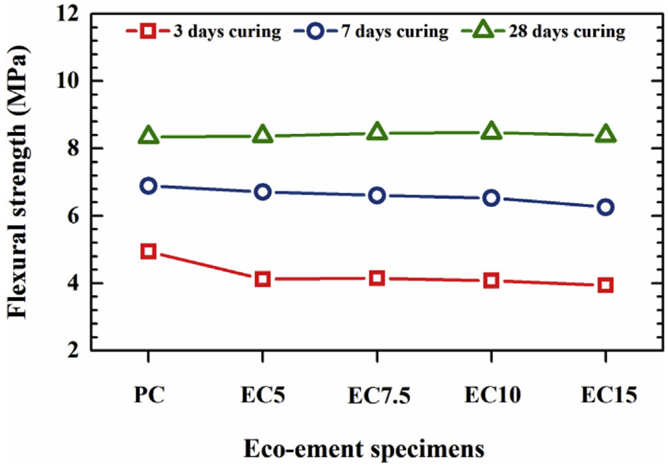

**Fig. 8.** The effect of different replacement levels of dry sewage sludge and curing periods on flexural strength of eco-cement specimens.

observed that the trend of compressive and flexural strength gains for all eco-cement specimens during first 28 days of curing were fairly similar to Portland cement, as mentioned by other researchers, too (Lin et al., 2012). The difference in strength development of eco-cement specimens compare with control ones in early curing times can attributed to the amount of $C_3S$ and $C_2S$ content which causes the delay to hydration reactions and subsequently setting time, as previously discussed. It especially can be noticed that 28-day strengths of all specimens are very similar. It seems like the effect of dry sludge helped and improve long-term strength of the eco-cement specimens (Lin et al., 2012). The first week compressive strengths of eco-cement specimens with higher levels of dry sludge displayed very small decline, most likely due to higher rate of hydration of finer cementitious material.

### *3.11. Volume expansion*

Some cracks and damages may appear in the cement paste if it expands. Therefore, cement should be manufactures in a way that the volume of its paste does not change after setting and during hardening. Probable cement paste volume expansions could occur due to the reaction of free lime, MgO, calcium sulfate, and other alkaline substances in cement. ASTM C150 standard has suggested some limitations on maximum allowable amount of free lime, MgO and other alkaline compounds.

The health of eco-cement pastes in this study were determined in accordance with ASTM standard test method using autoclave. Results have been presented in Fig. 9. It can be deduced that by increasing the dry sludge replacement level, the volume expansion of the cement paste increases. The reason could be attributed to the increase in the amount of free lime and MgO, as well as the total alkalinity. Nevertheless, at the highest replacement level of 15%, the volumetric expansion of cement paste was 0.159% and significantly less than the maximum permitted level specified in ASTM C 150 (0.8%).

## 4. Conclusion

This study was conducted to investigate the effects of using dry municipal sewage sludge as a partial replacement of raw materials in Portland cement production on physical, mechanical and durability properties of the eco-cement. According to the results obtained in this work, following conclusions could be made:

1. XRF results showed that all major chemical components of eco-cements (for all replacement levels of dry sludge) were the same or very similar to ordinary Portland cement. The presence of silicon, aluminum and iron oxides in the chemical composition of municipal sewage sludge makes this waste material suitable to substitute a portion of raw materials used in clinker production.
2. By replacing 15% of total raw materials in clinker production with dry sludge, the amount of bauxite, marl and iron ore consumption can be reduced by 50, 35 and 17%, respectively. Also, the amount of consumed lime will increase by 6%.
3. There are respectively direct and inverse relationship between the increase in replacement levels of dry sludge and $C_2S$ and $C_3S$ formation in eco-cement paste. The $C_2S$ and $C_3S$ formation depends on the availability of free lime in eco-cement. Furthermore, the amount of MgO with the highest sludge replacement level of 15% was equal to 3.07%, which is less than the maximum permitted level set in the standards. Also, the total alkaline contents of specimens with dry sludge replacement levels up to 7.5%, were lower than the standards value.

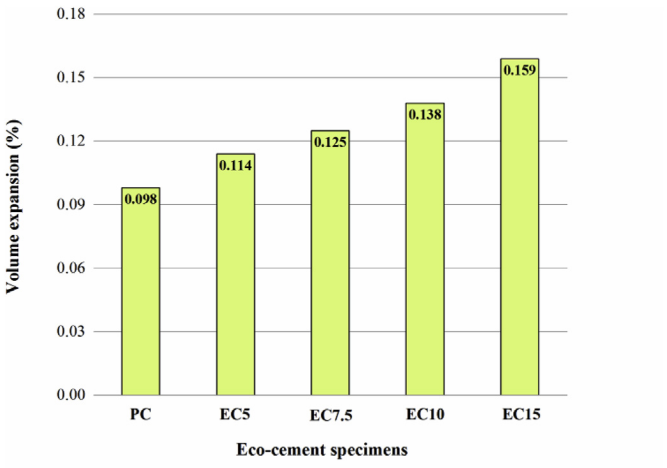

**Fig. 9.** The effect of various replacement levels of dry sewage sludge on the volume expansion of eco-cement.

4. With increasing the dry sludge replacement level, the specific gravity of the produced cements decreases due to incomplete combustion of organic matter left in the clinker cake or gas trapping caused by combustion of organic material in dry sludge. The former cause could also lead to an increase in the LOI value of produced eco-cement.
5. The amount of free lime in eco-cement specimens increases with increasing the dry sludge replacement level. Nevertheless, in the highest replacement level of 15%, the amount of free lime was 1.1% which is less than the maximum allowed value. It should be noted that increasing the amount of free lime in the eco-cement increases the fineness of produced cement. This could be contributed to reduction of energy consumption and increment of the clinker mill efficiency.
6. With increasing the dry sludge replacement level, both initial and final setting times increased due to declining of $C_3A$ and $C_3S$. Even at the highest replacement level of 15%, the setting times were below the maximum allowed value. More fineness and porosity in dry sludge containing clinker could result in higher amount of water needed to reach desirable consistency in concrete mixtures.
7. For all eco-cements, the 28-day mechanical strength would be approximately equal to that of ordinary Portland cement. Results show that the 7-day compressive strengths for eco-cements with 5, 7.5, 10 and 15% replacement levels were 4.6, 5.3, 9.4 and 17.2% lower than that of ordinary Portland cement, respectively.
8. It was noted that although the mechanical strength of the specimens decreases by increasing the dry sludge replacement level, however even in the eco-cement with 15% dry sludge replacement, 7-day compressive strength is higher than the minimum acceptable values.
9. Increasing in dry sludge replacement level leads to an increase in amount of volume expansion of the eco-cement paste. However, at the highest replacement level of 15%, the volume expansion was 0.159%, well below the maximum permitted level specified in the standard (0.8%).

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