Water-Retention Properties of Pavement Ceramics Prepared From Mixtures of Waste Diatomite and Light Emitting Diode Quartz Sand

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Abstract. This study investigated and analyzed the effects of the heating temperature (1,000°C-1,200°C), sintering time (6 hours), and content by weight (0-20%) of diatomite waste blended with a percentage of light emitting diode quartz sand (LEDQS) waste on fabricating water-retention pavement ceramics (WRPC). The results indicated that decreasing the heating temperature and amounts of the LEDQS resulted in an increase in the porosity. Increasing the heating temperature and amounts of the LEDQS waste resulted in the WRPC samples having increased compressive strengths (3.37-20.02 MPa). When amounts of up to 20% LEDQS were added to the WRPC samples heated to 1,200°C, the driving force was higher, and the crystal growth rate increased. Therefore, it is to be expected that densification will increase with increasing temperatures. It was found that the t₁/₂ values of the WRPC samples increased when the percentage of added LEDQS was increased. In summary, the WRPC samples containing LEDQS have excellent mechanical properties, making them feasible for use in WRPC applications.

1. Introduction
As the light-emitting diode (LED) market expands, large amounts of LED waste are being generated because of the limited lifespans and rapid update of LEDs used in electronic products [1]. Thus, LED quartz sand (LEDQS) waste is a crucial worldwide environmental concern. Treatment methods, such as landfilling and incineration, are not very effective because the stable structure and properties of the materials in end-of-life LEDs could not be altered. Diatomaceous earth is often used in food processing for filtering and eventually becomes diatomite waste [2]. Diatomaceous earth is typically soft, friable, fine grained, characterized by a relatively low density, chemically inert in most liquid and gases, and sparingly soluble in water with a low thermal conductivity [3]. Some minerals are considered to be raw materials appropriate for low cost ceramic products, as they are cheap and incur low fabrication costs because of their low sintering temperatures. Diatomite is an attractive material for the fabrication of porous ceramics due to its low cost, natural porosity and attractive properties [4, 5]. Several materials with water-retention properties have been developed for use in various types of functional pavement systems. The surface temperature is expected to be reduced since the water retained in the pavements consumes the heat around it when it is evaporated by water-retention
pavement ceramics [6]. Water-retention pavement ceramics (WRPC) can facilitate water cycling, thereby controlling climate change and preventing urban heat island effects. Some studies have prepared and characterized the properties of WRPC. However, the microstructure and characteristics of the porous ceramics obtained from diatomite and LEDQS waste have not yet been reported in the literature. The aim of this study is to investigate the effects of diatomite and LEDQS waste additions on the sintering properties of samples by examining the porosity, shrinkage, compressive strength, and water-retention performance, and X-ray diffraction (XRD) and scanning electron microscopy (SEM) techniques were determined to assess the suitability of diatomite and LEDQS waste additions for overcoming the urban heat island effects.

2. Materials and methods

2.1. Materials
The LEDQS used in this study was collected from an optoelectronics corporation in northern Taiwan. After the wasting process was performed by sand blasting, the wafer carrying platform produced LEDQS. The diatomite and LEDQS waste were pulverized using a ball mill until the resulting powder could pass through a 100 mesh (0.149 mm) sieve. The resultant pulverized diatomite and LEDQS waste was then stored in a desiccator until they were tested.

2.2. Preparation of compacted sintered WRPC samples
The diatomite and LEDQS waste samples were oven-dried at 105°C for 24 h and ground in a ball mill to form fine powders (until the powders could pass through a 100 mesh sieve) suitable for pressing. After drying, the mixtures were granulated in moist conditions and then semidyed pressed at 5 MPa to prepare specimens with 51.8 mm diameters and 15 mm thicknesses. The WRPC samples were heated to various temperatures (1,150°C, 1,100°C, 1,150°C and 1,200°C) for 2 h in a programmable electric furnace containing air at a ramp rate of 5°C/min and cooled down to room temperature naturally. Then, the sintered WRPC samples were cooled to room temperature in the furnace and stored in a desiccator for subsequent physical property testing and microstructural analyses. The open porosity measurement was performed on the sintered specimens using the Archimedes method. The XRD patterns of the WRPC samples were obtained by a Rigaku MiniFlex 600 diffractometer. The microstructure of the WRPC samples was observed using a Hitachi S-3500N SEM system. The water absorbed after 24 h was defined as the water absorption (Wa (%)). The release of the absorbed water was evaluated in an atmosphere with the relative humidity controlled at 55% using a saturated Mg(NO₃)₂·6H₂O solution. The water release rate was then evaluated from the time at which half the absorbed water was lost (t₁/₂ (h)) [7].

3. Results and discussion

3.1. Characteristics of diatomite and LEDQS waste
The LEDQS had a pH and specific gravity of 9.43 and 2.4, respectively. Table 1 presents the main constituents of the diatomite and LEDQS waste. The main constituents of the diatomite waste were 97.8% SiO₂, 0.87% Fe₂O₃, and 0.25% SO₃. Diatomite waste containing a large amount of SiO₂ can be a good raw material for WRPC. In addition, the main chemical composition of LEDQS was as follows: 69.1% SiO₂, 17.1% Na₂O, 9.1% CaO, 3.19% MgO, 0.28% Al₂O₃, 0.25% SO₃, and 0.11% Fe₂O₃. LEDQS, when incorporated into a mixture, has a good potential as a new fluxing agent to replace traditional feldspar during the sintering of WRPC.
Table 1. Chemical composition of raw materials.

| Composition | Waste Diatomite | LED Quartz Sand |
|-------------|----------------|-----------------|
| SiO$_2$ (%) | 97.8           | 69.1            |
| Al$_2$O$_3$ (%) | 0.58   | 0.28            |
| Fe$_2$O$_3$ (%) | 0.87     | 0.11            |
| CaO (%) | 0.18           | 9.07            |
| MgO (%) | -              | 3.19            |
| SO$_3$ (%) | 0.25           | 0.25            |
| Na$_2$O (%) | -              | 17.1            |
| K$_2$O (%) | 0.21           | 0.12            |

3.2. Mechanical characteristics of the WRPC samples

In this investigation, the porosity obtained during the sintering of the WRPC samples was considered. Fig. 1 shows the open porosity of the WRPC samples. When the amount of the LEDQS waste was from 0% to 40%, the porosity was 66.1%, 64.3%, 64.0, 62.6% and 59.4%. It is commonly recognized that the first stage of sintering at lower temperatures involves more complicated processes, such as the sintering of the agglomerated particles, which is driven by the decrease in the surface area and the neck-formation that occurs between the particles by both reactions and surface diffusion [8]. As shown in Fig. 1, for LEDQS contents from 10% to 40%, the porosity was from 60.8% to 64.1%, 57.5 to 63.0%, 54.0% to 60.6% and 52.5% to 57.6% when the heating temperatures were 1,000, 1,050, 1,100 and 1,200°C, respectively. This is possibly due to the increased LEDQS addition and cristobalite phase formation, which leads to the WRPC sample volumes decreasing during sintering, and most of the pores are closed, which improves the densification process [9].

Figure 1. Open porosity of the WRPC samples.

Figure 2 plots the shrinkage of the WRPC samples at various heating temperatures. The shrinkage of the pure diatomite WRPC samples was 3.39%, 4.84%, 7.07% and 8.03% upon heating to temperatures of 1,000, 1,050, 1,100 and 1,200°C, respectively. No significant shrinkage was observed below 1,000°C, and rapid densification occurred above 1200°C. When the LEDQS content in the mixture was varied from 5-20%, the shrinkage of the WRPC samples was 3.56%, 3.57%, 3.61% and 3.64%
upon heating to a temperature of 1,000°C. When the heating temperature was 1,000°C, a high energy barrier associated with the shrinkage process was observed at this stage. When the LEDQS in the mixture was varied from 5-20%, the shrinkage of the WRPC samples was 8.04%, 8.81%, 9.05% and 9.11% upon heating to a temperature of 1,200°C. Moreover, the shrinkage of the WRPC samples increased with increasing LEDQS contents. According to the sintering theory, the activation energy is essentially related to the mass transfer that occurs via both surface and grain boundary diffusion, which causes fast shrinkage.

Figure 2. Shrinkage of WRPC samples.

The compressive strength is a crucial index of the engineering quality of the WRPC samples [10]. Figure 3 shows the compressive strength of the WRPC samples. The compressive strength of the WRPC samples increased when the heating temperature increased from 1,000 °C to 1,200 °C. The compressive strength of the pure diatomite sintered WRPC samples was 2.39 MPa, 3.37 MPa, 4.09 MPa and 5.53 MPa upon heating to temperatures of 1,000, 1,050, 1,100 and 1,200°C, respectively. When the LEDQS in the WRPC samples was varied from 5-20%, the compressive strength of the WRPC samples was 3.37 MPa, 4.88 MPa, 10.51 MPa and 11.01 MPa upon heating to 1,000°C. When the LEDQS content in the WRPC samples was varied from 5-20%, the compressive strength of the WRPC samples was 8.15 MPa, 14.51 MPa, 15.92 MPa and 20.02 MPa upon heating to 1,200°C. The results showed that the optimal heating temperature that maximized the compressive strength was 1,200 °C. The compressive strength of the mixed WRPC samples containing the LEDQS waste increased obviously when the heating temperature increased above 1,200 °C. [11].
Figure 3. Compressive strength of WRPC samples.

3.3. Scanning electron microscopy microphotographs of the WRPC samples

Figure 4 (a) shows the SEM microphotographs of the WRPC samples containing 5% LEDQS sintered at 1,200 °C which involved more complicated processes, such as sintering within the agglomerated particles, which was driven by the increased surface area and the neck-formation that occurs between the particles by both reactions and surface diffusion. The rapid increase in the density of the WRPC samples containing 10% LEDQS at a heating temperature of 1,200 °C can be related to the formation of a melt phase, which promotes liquid phase sintering and generates numerous closed pores (Figure 4 (b)). The formation of a melt phase is corroborated by the micrographs of the WRPC samples containing 20% LEDQS (Figure 4 (d)), which was obtained from the fracture surface of the monoliths heated at 1,200 °C. Possible agents of fusion in the LEDQS include Na$_2$O, CaO, MgO, Al$_2$O$_3$, K$_2$O, and Fe$_2$O$_3$, which favor the formation of low temperature eutectics and, thus, the formation of a melt phase in the silica-rich grains [12].
3.4. XRD patterns of the WRPC samples

Figure 5 shows the XRD patterns of the WRPC samples sintered at 1,000 °C, 1,050 °C, 1,100 °C, and 1,200 °C. The main peak from the pure diatomite WRPC samples sintered at 1,000 °C show the formation of a cristobalite phase (2θ=21.4°, 27.8°, 30.8° and 35.6°). The cristobalite content increased as the temperature increased. The new peaks appearing at 1,100°C indicate that a cristobalite phase is being formed, and its content becomes higher with an increase in the temperature. This crystallized cristobalite phase is preferred over amorphous silica for many applications because of its greater chemical and thermal stability [14]. When the sintered temperature reaches 1,200°C, all the amorphous silicon dioxide is converted to the crystalline phase, and cristobalite becomes the major phase. The crystalline phase of the WRPC samples containing LEDQS did not change. This agree with the results presented by other investigators [14].

3.5. Water-release parameter (t_{1/2}) of the WRPC samples

Figure 6 shows the water-release parameter (t_{1/2}) of the WRPC samples. The water-release parameter of the pure diatomite WRPC samples was 6.1 h, 7.0 h, 7.1 h and 7.5 h upon heating to temperatures of 1,000, 1,050, 1,100 and 1,200°C, respectively. The water-release parameter increased as the temperature used to heat the pure diatomite WRPC samples increased. The water-release parameter increased from 6.1 h to 11.2 h, 7.0 h to 12.5 h, 7.1 h to 12.8 h, and 7.5 h to 13.1 h when the heating temperatures were increased to 1,000 °C, 1,050 °C, 1,100 °C, and 1,200 °C, respectively. Increasing the heating temperature from 1,000 °C to 1,200 °C improved the water-retention properties of the resulting WRPC samples containing LEDQS. When the heating temperature reached 1,200 °C, the WRPC samples containing 20% LEDQS had a t_{1/2} value of 13.1, which was larger than that obtained for the foamed glass sample of 4 h. The WRPC samples containing LEDQS exhibited excellent slow
water-releasing properties, which may be attributed to the pores being smaller than those in the foamed glass [7].

Figure 6. Water-release parameter (t1/2) of WRPC samples.

4. Conclusion
Based on the results of this study, no significant shrinkage was observed below 1,000°C, and rapid densification occurred above 1,200°C. Moreover, the shrinkage of the WRPC samples became more significant with increasing LEDQS contents. According to the sintering theory, the activation energy is essentially related to the mass transfer that occurs via both surface and grain boundary diffusion, which causes fast shrinkage. The optimal heating temperature that maximized the compressive strength was 1,200 °C. Cristobalite formation and physical sintering occurred at temperatures higher than 1,050°C. The rapid increase in the density at a heating temperature of 1,200 °C can be related to the formation of a melt phase, which promotes liquid phase sintering. The formation of a melt phase is corroborated by the micrographs of the WRPC samples containing 20% LEDQS, which were obtained from the fracture surface of the monoliths heated at 1,200 °C. The WRPC samples containing LEDQS exhibited excellent slow water-releasing properties, which may be attributed to the pores being smaller than those in the foamed glass. It is feasible to use the WRPC samples containing LEDQS for use as water-retaining materials to combat “heat island” effects.

5. References
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