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Utilization of Drinking Water Treatment Sludge as Cement Replacement to Mitigate Alkali–Silica Reaction in Cement Composites

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Abstract: Alkali–silica reaction (ASR) attack is one of the most significant durability concerns in cement-based materials. In this paper, the drinking water treatment sludge (DWTS), which is a typical by-product from the drinking water treatment industry, was reused as supplementary cementitious material to mitigate the degradation of mortar resulting from ASR attack. DWTS was milled and calcined at 800 °C for 2 h before being used as a replacement for cement. Glass sand was used as the reactive fine aggregate. Properties of four mortar mixtures prepared with 0%, 5%, 10%, and 20% of calcined DWTS replacement of cement were firstly assessed, including compressive strength, flexural strength, and water sorptivity. The mortar specimens were then exposed to an ASR-attacked environment for 28 days, the changes in specimen length were monitored, and the uniformity of mortar was measured via Ultrasonic pulse velocity (UPV). The results showed that 10% replacement significantly improved the mechanical properties of mortar. The specimens with 20% of the calcined DWTS exhibited comparable strength relative to the reference group and exhibited superior resistance to ASR attack. Additionally, a water sorptivity test showed that higher contents of the calcined DWTS can lead to lower water capillary absorption of mortar.

Keywords: drinking water treatment sludge; Alkali-silica reaction; cement replacement; mitigation

1. Introduction

Alkali–silica Reaction (ASR) has been one of the most common durability problems of concrete structures and causes significant concerns about structural integrity and service life. ASR is an expansive chemical reaction that occurs between the alkali ions in the pore solution from the cement paste and the amorphous silica in the aggregates when sufficient humidity is maintained [1,2]. The mechanisms of ASR damage can be divided into the following sequential reactions: (i) dissolution of amorphous silica, (ii) formation and gelation of colloidal silica, and (iii) swelling of the gel [3]. At the initial stage of ASR, the reactive silica is dissolved by OH^- attack, then the dissolved silica is cross-linked with the existence of Ca^{2+} [4,5]. As the reaction proceeds, the cross-linked silica and calcium coagulate form ASR gel. The ASR gel has a large surface area and contains hydrophilic groups, resulting in adsorption of water, expansion of the gel, and ultimately causing cracking of concrete. However, recent studies demonstrated the considerable existence of crystalline ASR products rather than amorphous ASR gel located inside cracks of aggregate. The crystalline ASR products have a layer-silicate structure similar



to mineral shlykovite [6]. Further study [7] exhibited that the crystalline ASR products could not swell by absorbing water and thus the hypothesis of gel expansion could be excluded from the attribution of cracking cement-based materials by ASR.

It has been found that the use of Supplementary Cementitious Materials (SCMs) such as fly ash, coal bottom ash, ground granulated blast furnace slag, silica fume, etc., could effectively mitigate ASR [8–11]. The ASR mitigation mechanisms related to the enrichment of silica and alumina in these SCMs can be summarized as: (i) replacing cement with SCMs reduces the Ca/Si ratio. The Calcium Silicate Hydrate (C-S-H) with a lower Ca/Si ratio has a stronger ability to bind alkali in pore solution than C-S-H provided from original cement hydration. This attributes to the increasing volume fraction of Si-OH on the surface of pozzolanic C-S-H [4,5,12–17]; (ii) The calcium hydroxide (CH) from cement hydration is consumed by the pozzolanic reaction and generates extra C-S-H which fills pores in the cement matrix, resulting in a lower rate of mass transportation for moisture and alkaline [18–21]; (iii) CH is considered as necessary for generating ASR gel and the consumption of CH by SCMs can reduce the possibility of forming ASR gel [22]; (iv) SCMs with higher alumina produce more calcium aluminosilicate hydrate (C-A-S-H) during pozzolanic reaction, which enhances the ability to bind alkali. Additionally, the aluminium incorporated into the surface of silica can significantly decrease the dissolution of amorphous silica [21,23–27].

Drinking Water Treatment Sludge (DWTS) is a by-product generated during water purification processes such as sedimentation and filtration. Owing to the growth of drinking water demand, the amount of DWTS has consequently risen. It was estimated that more than 342k tons of DWTS were produced in the years of 2017–18 in Australia [28]. After mechanical dewatering, most of the DWTS is dumped in landfill sites. Although DWTS can be considered as non-hazardous waste [29], the cost for landfill is significant. In Victoria, the annual cost to dispose of DWTS is over 6.2 million Australia dollars [30]. To save the cost for DWTS disposal, recycling the DWTS into construction materials is generating great interest.

The major chemical components of untreated DWTS are organic contents, alumina, and silica [31]. If the untreated DWTS is calcined at temperature higher than 600 °C, the organic contents will be significantly reduced to less than 10%, while alumina and silica will become the most abundant components in calcined DWTS. Therefore, based on the main chemical composition in the calcined DWTS and the ASR mitigation mechanisms, utilizing calcined DWTS as SCM has the potential to control the ASR-derived degradation of cement-based materials.

Recent studies have shown the utilization of DWTS to partially replace cement or fine aggregate and to produce blocks and ceramic products. Rodriguez et al. [32] prepared several mixtures of mortars in which cement was replaced by atomized DWTS at different contents varying from 10% to 30%. However, a significant drop in compressive and flexural strength was observed with the increasing replacement proportions of DWTS due to the high organic contents in the un-calcined DWTS. In addition, the study [33] found that the incorporation of un-calcined DWTS into concrete blocks deteriorated the durability of the resulting products, including abrasion and resistance to sulfate attack. Benlalla et al. [34] incorporated 5% to 30% of DWTS into clay to manufacture ceramic bricks. The compressive strength, bulk density, and water absorption of the ceramic bricks were evaluated from 800 to 1000 °C. The results revealed that 20% replacement of clay with DWTS led to the best mechanical properties. Yue et al. [31,33,35] studied the mechanical performance of the DWTS incorporated concrete blocks. The calcined DWTS was used as the fine aggregate replacement up to 30% and the blocks were produced through a dry mix method with similar water to binder ratio for each mixture. The authors reported that the compressive strength was improved by 7% when 5% of the natural sand was replaced by DWTS, and a comparable strength relative to that of the reference specimens was found when 10% of the DWTS was incorporated into the block specimens. According to the literature on the studies of sludge-derived products, the emphasis has already been put on the mechanical properties and some durability properties, while ASR resistance of cement-based composite incorporating calcined sludge was still not been researched.

In this study, the DWTS was milled and calcined to activate its pozzolanic activity. Mechanical and durability properties of sludge-derived mortar were evaluated to explore the optimal replacement ratio of cement by calcined DWTS. An accelerated ASR test was conducted in which cracking observation, expansion test, and ultrasonic pulse velocity test were taken to assess the effect of DWTS on ASR mitigation.

2. Materials and Methods

2.1. Materials

In this study, glass sand, which contained high contents of silica to activate much more serious ASR than natural sand [36], was used as reactive fine aggregates. The glass sand was provided by Schneppa Glass TM, Melbourne, Australia. The particle size distribution of the glass sand was measured by Malvern[®] MASTERSIZER 3000 (Malvern Instruments, Malvern, UK). Figure 1 shows the image and the particle size distribution of the glass sand. D90, D50, and D10 of the measured glass sand were 1580, 729, and 296 μ m, respectively. In addition, the binder used in this study was Type General Purpose Portland cement (GPC), conforming to AS 3972 [37].



Figure 1. (a) Image of the glass sand and (b) its particle size distribution.

The DWTS used in this study is from a local water treatment plant in South Australia. The chemical components of the cement, the untreated DWTS and the calcined DWTS were analysed by X-Ray Fluorescence Spectrometry (XRF) and the results are showed in Table 1.

Chemical Analysis (wt%)	GPC	Untreated DWTS	Calcined DWTS
Al ₂ O ₃	3.57	28.27	47.68
SiO ₂	20.78	26.43	31.11
Fe ₂ O ₃	3.99	7.66	4.94
CaO	61.8	5.36	4.32
K ₂ O	0.82	1.23	0.97
MgO	2.65	1.11	0.96
CuO	-	0.71	0.29
S/SO ₃	2.82	0.48	3.39
Na ₂ O	0.06	0.06	0.19
LOI	3.26	29.5	6.15

Table 1. Chemical composition of General Purpose Portland cement (GPC), untreated drinking water treatment sludge (DWTS), and calcined DWTS.

The major components of the untreated DWTS are alumina, silica, and Loss On ignition (LOI), occupying 84.2% of the total chemical composition. The abundant elements of aluminium and silicon imply the pozzolanic ability of DWTS. The large amount of LOI due to a high content of organic matter in the untreated sludge could significantly impede cement hydration. Additionally, the particle size of the DWTS could strongly influence the pozzolanic reactivity. Therefore, to remove the organic matter and to activate the pozzolanic activity, the untreated DWTS was further processed by milling and calcining.

The untreated DWTS was firstly oven-dried at the temperature of 105 °C, then milled to pass a 75 μ m sieve in which the size was similar to a cement clinker, before calcining for 2 h at 800 °C. The chemical components of the calcined DWTS illustrated in Table 1 have been analysed by XRF. Based on the result, the alumina and silica increased by 48% and 31%, respectively, while the total content of alkali metal oxide and alkali earth metal oxide decreased slightly after the volatile organic compounds were removed from 30% to only 6% through the calcination.

The particle size of the calcined DWTS and the cement are shown in Figure 2. The particle size distribution of the calcined DWTS is similar but slightly coarser than that of the cement. D90, D50 and D10 of the calcined DWTS are 53 μ m, 27 μ m and 5.5 μ m, respectively.



Figure 2. Particle size of the calcined drinking water treatment sludge (DWTS) and cement.

2.2. Mix Compositions and Mortar Preparation

Four mixtures were prepared according to the recommendation of AS 1141.60.1 [38]. For the controlled mixtures (M0), 440 g cement and 990 g sand glass were mixed with the water to binder ratio of 0.47 by weight. The other three mixtures were designed by replacing cement from 5% to 20% with the calcined DWTS, annotated as M5, M10, and M20, respectively. The detailed mixture proportions are shown in Table 2. Due to the significant water absorption capacity of the calcined DWTS, to ensure the fresh sludge-cementitious mixture having a similar flowability (approximately 150 mm in diameter) to that of the control specimens, superplasticizer complied with ASTM C 1437 [39] was introduced.

Specimen Notation	Cement (g)	Calcined DWTS (g)	Glass Sand (g)
M0	440	0	990
M5	418	22	990
M10	396	44	990
M20	352	88	990

Table 2. Mixtures proportions used for preparing specimens.

As for mixing processes, the GPC and the calcined DWTS were firstly mixed for 10 min to ensure a homogenous binder system. Glass sand was then added and mixed for an extra 1 min. In the final phase, water and superplasticizer were premixed together and poured into received mixtures for another 2 min. For each mix, 6 flexural specimens ($40 \text{ mm} \times 40 \text{ mm} \times 160 \text{ mm}$) (3 for flexural tests and 3 for ASR test) and 3 cubic specimens ($50 \text{ mm} \times 50 \text{ mm} \times 50 \text{ mm}$) (for compressive test) were poured. As for the flexural moulds used for making the ASR specimens, two stainless steel gauge studs were attached at the ends before placing in the cementitious mixture. All mortar bars were demoulded right after 24 h of initial curing. With regard to specimens used for assessing the fundamental mechanical properties (compressive and flexural strength), the specimens were immersed in a water tank, which was stored in a temperature and humidity-controlled room (50 °C and 50%) for a period of 28 day curing.

2.3. Compressive Strength Test

A compressive strength test was undertaken referring to AS 4456.4 [40]; three cubic specimens for each mixture were tested by the Compression Testing machine with a loading rate of 0.33 MPa/s until failure.

2.4. Flexural Strength Test

Flexural strength of the mortar specimens was determined according to ASTM C 293 [41]; three beam specimens for each mixture were tested by the Flexure Testing machine with a loading rate of 0.5 kN/s.

2.5. Water Sorptivity Test

Water sorptivity was tested based on ASTM C 1585 [42]. The mortar specimens were oven-dried at 40 °C until the weight difference between two successive values was less than 0.5% of the less value. The side surfaces were then sealed with epoxy and the top surface was attached with plastic bags to avoid evaporation. The specimens were placed into containers where the water level was kept constant to be 3 mm above the bottom surface of the specimens. The weight of the specimens was measured at 60 s, 5 min, 10 min, 20 min, 60 min, then every hour up to 6 h in the first day and once a day for 9 days. The absorption depth *I* (mm) is calculated by Equation (1), where m_t is the mass in gram at time *t*, m_0 is the initial mass before the specimen contacts with water, *a* is the area of the contact surface in mm², and *d* is the density of water in g/mm³.

$$I(mm) = \frac{m_t - m_0}{a \times d} \tag{1}$$

2.6. Accelerated ASR Test

Accelerated Mortar Bar Test (AMBT) via AS 1141.60.1 [38] was used to evaluate whether incorporation of the calcined DWTS could help to mitigate the ASR effect of the glass–sand mortar. The mortars in moulds were covered by plastic sheets and placed in a curing room fixed at a temperature of 23 °C and a relative humidity of 50% for 24 h. Afterwards, specimens were demoulded and immersed in a water bath in which the temperature was raised from 23 °C to 80 ± 2 °C in 2 h and the temperature was maintained for another 22 h. Zero readings were then carried out to obtain the initial length and the ultrasonic velocity of the mortars. After curing in the heated water for 24 h, the specimens were immersed in a container filled with 1 mol/L (1M) NaOH solution and the temperature of the solution was maintained at 80 ± 2 °C.

The length of the specimens under ASR attack was measured at the initial, 3rd, 7th, 14th, 21th, and 28th day using a length comparator, and the expansion percentage is determined as Equation (2), where L_i is the length at the *i*th day, L_0 is the initial length of the specimen.

$$Expansion (\%) = \frac{L_i - L_0}{L_0} \times 100$$
⁽²⁾

Ultrasonic pulse velocity (UPV) was measured to evaluate uniformity mortars under the ASR attack according to ASTM C 597 [43]. A frequency of 54 kHz was used and the transmitting time was recorded. UPV is calculated based on the transmitting time and the distance between the sensors. For each mortar bar, at least five measurements were taken to limit errors with 1% of the mean value. UPV of specimens was measured on the initial day and the 28th day after immersion in the NaOH solution.

3. Results

3.1. Compressive and Flexural Strength

Table 3 shows 28-day compressive and flexural strengths of mortar specimens. When 5% and 10% of the cement are replaced by the calcined DWTS, the compressive strength is 44 MPa and 46 MPa, respectively. Thus, significant improvement of the compressive strength was observed for M5 and M10 compared with that of the reference specimens (35 MPa). Such improvement was not found for M20, in which the value of compressive strength is 33 MPa, but it is still comparable with that of the reference specimens.

Specimen Notation	Compressive Strength (MPa)	Flexural Strength (MPa)
M0	35 (±3.8)	5.41 (±0.11)
M5	44 (±1.9)	5.82 (±0.29)
M10	46 (±5.5)	6.18 (±0.51)
M20	33 (±2.8)	5.56 (±0.22)

Table 3. Compressive and flexural strengths of the mortars with 0%, 5%, 10%, and 20% cement replaced by the calcined DWTS.

The flexural strengths of the mortars M0, M5, M10, and M20 were 5.41 MPa, 5.82 MPa, 6.18 MPa, and 5.56 MPa, respectively. The trend is similar with the compressive strength. The maximal flexural strength appeared when 10% of cement was replaced by the calcined DWTS. As for mortars incorporating more than 10% of the calcined DWTS content, the flexural strength began to decrease but was still higher than that of the reference sample.

3.2. Water Sorptivity

The results of the water sorptivity are shown in Figure 3. The reference specimens had the highest capacity of water capillary absorption, followed by M5 and M10, while M20 had the lowest ability to absorb capillary water. It can be seen that with the increasing proportion of cement replacement by the calcined DWTS, the water sorptivity of the specimens was decreasing which means an improvement of durability. Additionally, the slopes of the curves during the initial absorption (before the first 6 h) were reducing when the DWTS contents were increasing in the mortar.



Figure 3. Water sorptivity of the mortars with 0%, 5%, 10%, and 20% cement replaced by the calcined DWTS.

3.3. ASR Assessment

3.3.1. Visual Observation of Cracking under Accelerated ASR Test

Figure 4 shows the cracking images of the specimens after 28 days test. Dense and wide cracks were observed for M0 and M5. As for M10, cracks were sparser and thinner than that of M0 and M5. No obvious cracks can be seen for M20.



Figure 4. Visual observation of mortar cracking after 28 days immersion in 1M NaOH solution.

3.3.2. Expansion under Accelerated ASR Test

Figure 5 shows the expansion ratios of the specimens. In the first 3 days, the negative values of expansion were obtained for all tested specimens, indicating the chemical shrinkage due to the cement hydration. After 3 days, positive expansion could be observed. Small expansion was observed for all mortars until the end of 14th day, while dramatically increased expansion appeared after 14 days test for M0 and M5. M10 started to show a clear expansion after 21 days. Expansion for M20 was slightly increasing and tending to be stable after 21 days.



Figure 5. Expansion of the mortars with 0%, 5%, 10%, and 20% cement replaced by the calcined DWTS.

3.3.3. UPV changes under accelerated ASR test

The UPV results are shown as Figure 6. The dashed line in the figure shows the absolute values of the relative differences between the initial day and the 28th day. The initial UPVs were almost the same for M0 and M5. M10 and M20 exhibited slightly lower values of UPVs than that of M0 and M5. After 28 days, the UPVs of M0 and M5 decreased to 2.64 km/s, which was approximately 43% lower than that of the initial values. As for M10, only a 19% decrease of UPVs was shown, which signaled the effort of the calcined DWTS to mitigate ASR. Furthermore, the UPV for M20 increased by 19% compared with that of the initial UPVs after 28 days test. This could be explained by the fact that the formed aluminosilicate had accommodated some void in the mortar, reducing the void proportion and thus increasing the UPV.



Figure 6. UPV at initial and 28 days of the mortars with 0%, 5%, 10%, and 20% cement replaced by the calcined DWTS.

4. Discussion

4.1. Effect of the Calcined DWTS on Mechanical Properties

The improvement of the compressive and flexural strength could be explained by the pozzolanic activity from calcined DWTS. The calcined DWTS reacted with CH which had been generated through the cement hydration and formed extra C-S-H in the binder matrix, therefore improving the mechanical performance of the mortar [44]. With 10% of the cement replaced by the calcined DWTS, the total amount of C-S-H including that generated from cement in the primary hydration and from the calcined DWTS in the secondary hydration could reach the optimum, leading to a maximum of compressive and flexural strength. When 20% of cement was replaced by the calcined DWTS, only limited C-S-H was generated in the primary cement hydration because the amount of CH, which could slow down the formation of the C-S-H generated via pozzolanic reaction in the secondary hydration.

4.2. Effect of the Calcined DWTS on Durability Performance

With an increase in the content of the calcined DWTS from 5% to 20%, the water sorptivity decreased. The results indicated that the pore size refinement from the pozzolanic reaction of the calcined DWTS was significant. With more contents of the calcined DWTS incorporated into the cement, more C-S-H could be generated from the pozzolanic reaction between the calcined DWTS and CH. The extra formed C-S-H may fill the large and open pores, converting them to gel pores or closed pores, thereby reducing the pore connectivity. Therefore, lower water capillary absorption of the mortar specimens was observed compared to those with open pores.

Interestingly, these results were not consistent with corresponding results from compressive and flexural strength tests. Generally, the denser structure should contribute to higher strength. But for the calcined DWTS, the structure of the un-reacted DWTS seems to weaken the skeleton of the composite matrix, resulting in a deteriorated mechanical performance [31]. Thus, the harness of inclusion should be regarded as an important characteristic to assess the mechanical properties of mortar specimens as well.

4.3. Effect of the Calcined DWTS on Mitigating ASR

The visual observation of cracks on the surface of the specimens under an accelerated ASR test reflects that increasing replacement ratios of cement with the calcined DWTS up to 20% can effectively mitigate ASR. These results were consistent with the outcomes obtained from the expansion test. According to AS 1141.60.1 [38], if the expansion ratio at the 21st day is less than 0.1%, the aggregate can be defined as non-reactive material for ASR; if the expansion ratio after 10 days is less than 0.1% and after 21 days is between 0.1 and 0.3%, the aggregate can be defined as slow-reactive. As mentioned above, glass sand was a highly reactive material for ASR. Hence, it can be concluded that incorporating the calcined DWTS can effectively impede the degradation of ASR. Additionally, according to the previous discussion, the water sorptivity was reduced by incorporating the calcined DWTS. This observation indicated that the permeability of the mortar was decreased. Mortars with lower permeability would be more beneficial to resist ASR due to the fact that moisture will be difficult to transport into the cement matrix, preventing water absorption and the expansion of ASR gel.

The mitigation of ASR by DWTS could also be due to the consumption of CH by the pozzolanic reaction. The result of the enhanced mechanical properties by the calcined DWTS confirmed the formation of the additional C-S-H from the pozzolanic reaction. Simultaneously, the fraction of CH in mortars was reduced by the pozzolanic reaction. As CH is essential to form ASR gel, the reduced CH mitigated ASR in the specimens with calcined DWTS.

On the other hand, although increasing expansion occurred for the specimens with 5% and 10% of cement replaced by the calcined DWTS after 15 days, the delay before such rapid expansion was observed from the result of the expansion test. This was probably caused by the aluminium in the pore solution which slowed down the dissolution of amorphous silica. According to Chappex et al. [25] and Hay et al. [26], low concentration of aluminium in pore solution could reduce the silica dissolution. Due to the low concentration of aluminium in pore solution, it will be quickly depleted without a solid source of aluminium to replenish it. In this paper, the specimens were immersed in 1M NaOH solution where the excess amount of alkaline kept consuming the aluminium. The calcined DWTS in the mortar functioned as a reservoir to provide aluminium into the pore solution. The aluminium source in the mortars with 5% and 10% cement replaced by the calcined DWTS may not be able to maintain the concentration of aluminium in the pore solution after 15 days of immersion in 1M NaOH solution. In addition, the mass transport of aluminium from the calcined DWTS to the pore solution might be lower if a lower proportion of the DWTS was incorporated into the mortars. Although ASR was decelerated in these two specimens, the expansive ASR products could still be slowly formed and eventually cracked the mortars, creating more surface for the alkaline to attack, leading to a sharp increasing of expansion for these two specimens.

These mitigation mechanisms targeted at different stages of ASR. The dissolved aluminium released into the pore solution from the calcined DWTS could control the dissolution of silica, which contributes to preventing ASR in the early stage. The CH consumption by the pozzolanic reaction from the calcined DWTS could limit the formation of ASR products by reducing the reactant. Furthermore, the pore refinement caused by the calcined DWTS could suppress the transportation of water into the ASR products, confining the water absorption of ASR gel. However, the relative contribution of silica dissolution control, CH consumption, and pore refinement were not independently evaluated in the current paper and should be investigated in future research.

5. Conclusions

In this study, the aluminium and silicon based DWTS was incorporated into mortars as SCM after DWTS was treated by milling and calcining. The mechanical and durability properties, as well as the resistance of mortars incorporating calcined DWTS to ASR attack, were studied. From the test results, the following conclusions can be drawn:

- (1) The compressive strength and flexural strength of the mortars were improved when no more than 10% of the cement was replaced by the calcined DWTS in the composite. The optimal mix was 10% replacement, where the highest compressive strength and flexural strength of mortar were achieved.
- (2) The calcined DWTS was found to decrease the water capillary absorption of the mortars due to the large and open pores of the composites being confined by extra C-S-H formation from the pozzolanic reaction of the calcined DWTS. This confirms that inclusion of calcined DWTS could improve the durability of cementitious material.
- (3) Incorporating calcined DWTS into cementitious materials was able to mitigate the ASR effect. A 20% replacement of cement with the calcined DWTS appeared to successfully prevent ASR occurrence. The ASR mitigation mechanisms could be attributed to the formation of secondary C-S-H, the consumption of CH, the silica dissolution control by aluminium, and the reduction of permeability.

Utilization of DWTS, a by-product of drink water treatment, as SCM improved the mechanical and durability properties of concrete including resistance to ASR attack. It also contributes to the sustainable development of a circular economy and to a clean environment.

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