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ARTICLE



Effect of Alkaline Electrolyzed Water on Performance Improvement of Green Concrete with High Volume of Mineral Admixtures

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ABSTRACT

The strength and durability of concrete will be significantly reduced at high volume of mineral admixture, and the poor early strength of concrete also still needs to be solved. In this investigation, a highly active alkaline electrolyzed waters was used as mixing water to improve the early strength and enhance the durability of green concrete with high volume mineral admixture, the influences of alkaline electrolyzed water (AEW) on hydration activity of mineral admixture and durability of concrete were determined. The results showed that compared with natural tap water, AEW can accelerate early hydration process of cement in concrete and produce comparatively more hydrated products, leading to a 13.6% higher compressive strength than that of ordinary concrete at early age, but the improvement effect of AEW concrete was relatively reduced at long-term age. Meanwhile, the activity of mineral admixtures could be stimulated by AEW to some extent, the strength and durability performance of AEW concrete after double doping 25% slag and 25% fly ash can still reach the level of ordinary cement concrete without mineral admixtures. The SEM micromorphology of 7 d hydrated natural tap water cement paste was observed to be flaky and tabular, but the AEW cement pastes present obvious cluster and granulation phenomenon. The SEM microstructure of AEW concrete with mineral admixtures is more developed and denser than ordinary tap water concrete with mineral admixtures. Therefore, the AEW probably could realize the effective utilization of about 50% mineral admixture amount of concrete without strength loss, the cement production cost and associated CO_2 emission reduced, which has a good economic and environmental benefit.

KEYWORDS

Alkaline electrolyzed water; durability improvement; green concrete; mineral admixture; micromorphology

1 Introduction

The attendant massive pile of industrial waste not only polluted environment but also caused the waste of resources with the global process of industrialization, the large amount of industrial waste residue is used for concrete as mineral admixture, which can fully meet the requirements of the sustainable development of human society [1]. At present, high performance green concrete has been widely used in modern concrete structures, which is characterized by the use of mineral admixtures, such as fly ash and ground granulated blast furnace slag [2–5]. Considering that using mineral admixtures to replace part of cement can substantially decrease the hydration temperature rise of concrete and reduce the cracking risk of concrete



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[6,7], maximizing the use of mineral admixture under the premise of ensuring the high performance of concrete can promote the mechanical properties and durability of concrete and achieve the purpose of reducing cement, saving resources, green and environment friendly [8]. However, a large amount of researches showed the small percentage of singly-doped mineral admixture is beneficial in optimizing the workability and low cost but it may not improve the durability to any considerable extent [9–12], and the strength and durability of concrete will be significantly reduced at high volume [13–15], Faiz et al. [16] reported the reduction of 28 d compressive strength of concrete was 15.82%, 19.34% and 48.99% with the increase of 50%, 60% and 70% fly ash, respectively. By comparison, although multi-doped mineral admixtures can increase the replacement ratio to a certain extent, improve concrete workability and pumpability [17], the strength and durability of concrete with high volume mineral admixture is worse than that of ordinary concrete [18–20]. More important is that low early activity of mineral admixture and poor early strength of concrete still need to be solved [21].

The alkaline electrolyzed water (AEW) is a kind of highly active water, which has been widely used in food hygiene, environmental cleaning, agricultural industries, etc. [22–24]. Sun et al. [25] researched that mixing alkaline water can decrease the total porosity of concrete and increase the strength by 21%. Kimura et al. [26] showed that electrolyzed water can decrease the amount of maximum radiation ray rate by up to 10% than ordinary water mortar. Mandal et al. [27] indicated that electrolyzed water can reduce the initial and final setting time of cement. Chakraborty et al. [28] revealed the electrolyzed water in can enhance the mechanical properties of cement mortar at the fresh and early hardened states. In this investigation, a kind of AEW was used in green concrete with high volume mineral admixture as mixing water, which is expected to be beneficial not only for promoting the early hydration of cement, improving the early and late strength, enhancing the durability of concrete, but also it can be considered as a greener admixture for minimizing the cement consumption and construction cost, which can provide a new way and reference for the engineering application of concrete with high volume of mineral admixtures.

2 Experimental Programs

2.1 Experimental Materials

The ordinary Portland cement (P·O 42.5) was provided by Shanlv Cement Co., Ltd. (Qingdao, China) in this study, which met the requirements of "PRC standards for Portland cement and ordinary Portland cement" (GB 175-2007). The density is 3.16 g/cm^3 , the surface area is $3350 \text{ cm}^2/\text{g}$, the fineness is 2.3%. The S95 grade granulated blast furnace slag (GGBFS) was used in this study, the specific surface area $>400 \text{ m}^2/\text{kg}$, the density is 2.88 g/cm^3 , loss on ignition is less than 1.0%, meeting the requirements of "Ground granulated blast furnace slag for cement, mortar and concrete" (GB/T18046-2017). The I-class fly ash was provided by Weifang Huadian Co., Ltd., Weifang, China, the density is 2.24 g/cm^3 , water requirement ratio is 0.95, loss on ignition is 1.1%, which meets the requirements of "Fly ash used for cement and concrete" (GB/T 1596-2017), the XRF results of different mineral admixtures are shown in Tab. 1. The natural river sand was used as fine aggregate, crushed quartzite stone were used as coarse aggregate with 5–31.5 mm continuous grading. The detailed parameters of fine and coarse aggregates are shown in Tabs. 2 and 3, respectively. The NC-J type polycarboxylate superplasticizer was used, which is produced by Shandong Academy of Building Science, the water-reducing ratio was 28% at 2.0-2.2% dosage of the cement weight, the ratio of bleeding rate is less than 30%, and the content of chloride ion is not more than 0.06%.

Table 1: Chemical composition results of different mineral admixtures by XRF (%)

Туре	CaO	SO_3	SiO ₂	Al_2O_3	MgO	Fe ₂ O ₃	CO_2	TiO ₂	Others
Portland Cement	52.71	2.60	19.9	6.41	4.60	2.83	9.50	0.40	1.05
I-class Fly ash	8.20	1.40	48.8	25.2	1.30	6.50	3.90	1.60	3.10
Blast furnace slag	36.40	2.00	29.1	14.3	8.90	0.30	5.60	1.30	2.10

Fineness module	Specifications	Stacking density/(kg·m ³)	Apparent density/	Void ratio/%	Micro powder content/%	Mud content/	Crushing index/%
			(kg·m³)			%	
2.40	II class sand	1450	2590 40		1.00	0.70	13.1

 Table 2: Performance index of natural fine aggregate in this study

Water absorption/%	Moisture content/%	Content of needle-like particles/%	Crushing Stack index/% densi / (kg	kingApparent densityity $/(kg \cdot m^3)$ $g \cdot m^3$)	
1.70	0.42	4.05	11.2 1460	2510	

 Table 3: Performance index of natural coarse aggregate in this study

2.2 Production and Preparation of AEW

A kind of highly active AEW was used as mixing water to prepare concrete with high volume of mineral admixtures in this study. The AEW was prepared by Boxin BX-SQJ series fully automatic ion-exchange diaphragm alkaline electrolytic water equipment, as shown in Fig. 1. Pure water was electrolyzed to avoid the influence of impurities in water on experimental results, potassium carbonate (AR) was used as electrolyte, the concentration of K_2CO_3 electrolyte is 0.05%, the voltage of electrolysis is 380 V 50 Hz, the water inlet temperature: 10–30°C, the total inlet water flow was 32.5 L/H, the inlet alkaline water flow was 20 L/H, the electrolysis's time was 15 min, the pH of AEW was about 9.8 at this time. By adjusting the current, voltage, influent flow and electrolysis time, the AEW with different performance can be prepared. Under the condition of constant voltage, the concentration of K_2CO_3 electrolyte and current value should be increased when pH value increased.



Figure 1: Diaphragm type electrolytic cell

In an electrolyzing cell with diaphragm, due to K_2CO_3 electrolyte solution of definite concentration of electrolytic treatment, KOH is generated on the cathode, the water is decomposed into hydrogen and hydroxyl ions; meanwhile, carbonic acid is generated on the anode, the H₂O is decomposed into oxygen and hydrogen ions, it is an acid solution [29]. The schematic diagram of electrolysis principle is shown in Fig. 2. Ordinary tap water was used as mixing water for blank comparative analysis, the pH meter and ORP meter are shown in Figs. 3 and 4, which were used to test the pH and ORP values of AEW in this

study, ORP is the abbreviation of redox potential, which represents the relative oxidation or reduction degree of AEW [30], the detailed performance index of AEW is shown in Tab. 4.



Figure 2: Schematic diagram of electrolysis



Figure 3: pH meter for electrolyzed water



Figure 4: ORP meter for electrolyzed water

Туре	Ideal pH range	pH measured value	ORP measured value	Abbreviations
Ordinary tap water	7	7.51	343	РТ
K ₂ CO ₃ alkaline electrolyzed water	9~10	9.82	191	DJ

Table 4: Performance index of different mixing water in this study

2.3 Mix Proportions Design

Tab. 5 presented the detailed mix proportions of different concrete with mineral admixture in this study. The cement content is 420 kg/m³, the actual water consumption is determined by controlling the slump of concrete mixture in the range of 180–220 mm, the sand coarse aggregate ratio (s/a) was designed as 50%. The proportion of total mineral admixtures to the total cementitious materials amount in concrete is designed as 0%, 40% and 50%, respectively, and the proportion of fly ash (FA) and blast furnace slag (BFS) is 1:1 by mass. The ordinary tap water concrete is blank control group, and the marks are A1, A2 and A3 in turn, the corresponding marks of AEW concrete are B1, B2 and B3, respectively. The dosage of water reducer admixture to the cement amount was 2.0%–2.2%, the water reducing ratio is 28%–30%.

Table 5: Mix proportion design of different concrete with high volume of mineral admixture

Туре	Mark	С	BFS	FA	S	G	W	Ad
Natural tap water	A1	420	0	0	912	877	177	8.4
	A2	255	85	85	917	881	159	12.7
	A3	215	108	108	918	882	149	15.0
Alkaline electrolyzed water	B1	420	0	0	912	877	177	8.4
	B2	255	85	85	917	881	159	12.7
	B3	215	108	108	918	882	149	15.0

Note: C: cement; S: sand; G: stone; W: water; Ad: water-reducer admixture.

2.4 Experimental Methods

AEW and ordinary tap water were used to prepare different concretes with high volume of mineral admixtures. The slump of different concrete mixture was controlled in the range of 180-220 mm. According to "Standard for test method of mechanical properties of ordinary concrete" (GB/T 50081-2002) in China, three identical cubic concrete specimens were made into 150 mm \times 150 mm \times 150 mm for each specimen design. After demoulding, the concrete specimens are moved to the standard curing laboratory with the temperature of $(20 \pm 2)^{\circ}$ C and the relative humidity of $(90 \pm 5)^{\circ}$, the compressive strength tests of specimens are carried out at different age of 3, 7, 14, and 28 d, respectively. Moreover, according to "Standard for test methods of long-term performance and durability of ordinary concrete" (GB/T 50082-2009), the chloride ion permeability test for concrete was conducted through the RCM method [31], the penetration coefficient of Cl⁻ in concrete can be calculated out. The carbon dioxide concentration in the carbonization chamber was adjusted to (20 ± 3) %, the humidity was controlled at $(70 \pm 3)^{\circ}$ C and the temperature was controlled at $(20 \pm 2)^{\circ}$. The carbonation depth tests for different concretes were conducted to 3, 7, 14, 28 and 91 d [32]. Considering the factors such as test period and test conditions, the quick freezing method is used in the freeze-thaw cycle test of concrete. The concrete specimens were cured in the standard curing room until the age of 28 d, the mass loss of the test specimens was measured once every 25 freeze-thaw cycles, and the transverse fundamental frequency was measured by dynamic modulus tester. The accuracy of relative dynamic modulus and mass loss rate in the test results are determined to be 0.1%. The X-ray diffraction analysis test was performed to characterize the mineral composition of cement paste in concrete at 7 d by XRD device (D8 Advance, Bruker AXS, Germany), the micromorphology of the different concrete was observed by scanning electron microscopy (SEM), and the microstructure of different concrete with high volume mineral admixture can be characterized by XRD and SEM spectra, the influence mechanism of AEW on the performance of high volume concrete can be analyzed eventually.

3 Results and Discussions

3.1 Workability of Different Series of Concrete

The slump testing of concrete was given in Fig. 5, from Fig. 6, it can be seen that the under the condition of the same slump range (180–220 mm), the water consumption of AEW concrete are all lower than those of natural tap water concrete at same proportioning. For ordinary cement concrete without mineral admixture, the water consumption of B1 AEW concrete reduced by 5 kg/m³. This is likely that AEW can accelerate hydration process and setting of cement, which is mainly governed by early dissolution of cement grains and rapid flocculation of hydrated products in presence of OH ions [33], as shown in Eqs. (1) and (2). Meanwhile, the highly active AEW small molecular group with negative charge could be absorbed on the surface of cement particles, which can make cement particles with same charge evenly disperse due to electrostatic repulsion and release more combined water in concrete [34], leading to a workability improvement of AEW concrete than that of natural tap water concrete. Behnam et al. [35–37] showed that using magnetized water instead of tap water leads to a higher slump flow and a lower viscosity of concrete due to high activity and small water molecular group after magnetization, which is similar to functional mechanism of AEW in concrete.



Figure 5: Slump testing of concrete

At the same time, at the early age, KOH in electrolyzed water was likely to react with the unhydrated $CaO \cdot Al_2O_3$ in cement to produce potassium aluminate (a kind of condensation promoter), as shown in Eq. (3), which could promote the hydration rate and setting of cement, having a positive effect in improving the workability of concrete.

$$3CaO \cdot SiO_2 + nH_2O = xCaO \cdot SiO_2 \cdot yH_2O + (3 - x)Ca(OH)_2$$
(1)

$$CaO \cdot SiO_2 + 2KOH = K_2SiO_3 \cdot CaO + H_2O$$
⁽²⁾

$$CaO \cdot Al_2O_3 + 6KOH = 3K_2O \cdot Al_2O_3 + 3Ca(OH)_2$$
 (3)

In addition, the water consumption of natural tap water and AEW concrete both decreased with the increase of mineral admixture content, which is mainly affected by the content of water reducer admixture.



Figure 6: Water consumption of different concrete

3.2 Compressive Strength Development of Different Series of Concrete

From Fig. 7, it can be seen that the strength development of three AEW concretes at each curing age were all higher than those of natural tap water concrete at same proportioning, of which the strength improvement effect of B1 concrete was the best. Meanwhile, the strengths of natural tap water and AEW concrete both decreased with increasing mineral admixture content at each age. At the early age (3 d), the strengths of B1 and B2 AEW concrete increased by about 13.6% and 7.3% than that of A1 natural tap water concrete, and the strength of B3 concrete was slightly higher than that of A1 concrete. Sumit et al. [28] indicated a 16% higher compressive strength of electrolyzed water mortar than that of normal mortar at 3 d, Wang et al. [38] also showed a 20% strength improvement of electrolyzed water mortar, which is essentially in agreement with results in this study.



Figure 7: Compressive strength development of different series of concrete with curing age

In contrast, the strength growth rate of B1 and B2 AEW concrete at 28 d decreased significantly, increased by only 4.2% and 2.5% than that of A1 concrete, the strength of B3 concrete was close to that of A1 concrete, but the strengths of these three AEW concretes were obviously higher than those of A2 and A3 concrete. It showed that introducing the AEW in concrete can accelerate cement hydration and activate the activity of mineral admixtures, leading to an increase of strength at early age. However, at the long-term age, the cement hydration was also relatively sufficient, the activity and electrostatic repulsion of AEW decreased, the strength improvement effect of AEW concrete reduced.

3.3 Free-Thraw Cycles of Different Series of Concrete

From Fig. 8 and Fig. 9, the results showed that after 300 times freeze-thaw cycles, the relative dynamic elastic modulus loss and the weight loss rates of ordinary tap water concrete series was more evident than those of AEW concrete series. With the freeze-thaw cycles number increase, the declining rate of weight loss and relative loss modulus of AEW concrete series were slowed obviously, showing a better frost resistance, which can well meet the actual engineering requirements. Meanwhile, with the increasing of mineral admixture content in concrete, the freeze and thaw resistance of ordinary tap water concrete can still reach the level of A1 ordinary concrete after double doping 25% slag and 25% fly ash, the relative dynamic elasticity modulus and weight loss ratio were 91.4% and 1.6%, which was superior to those of A2 and A3 ordinary tap water concrete.



Figure 8: Relative dynamic elasticity modulus of concrete

3.4 Chloride Ion Penetration Property of Different Series of Concrete

The testing device for chloride permeability of different concrete is shown in Fig. 10, NJ-RCM Chloride diffusion coefficient tester for concrete, Beijing Naijiu Technology Co., Ltd., China, after standard curing, the cylinder concrete specimens were submerged in an ultrasonic bath for 15 min, then the positive and negative electrodes of specimens were immersed in the 0.2 mol/L KOH solution and 0.2 mol/L KOH solution containing 5%NaCl, respectively. The electric time was determined according to the initial electric current. The penetration coefficient of chloride ions of concrete was calculated by the penetration depth of chloride ions. The Cl⁻ penetration coefficients of ordinary tap water concrete and AEW concrete at different curing age are given in Fig. 11. From Fig. 11, it can be seen that the Cl⁻ penetration coefficients of three kinds of AEW concrete was the lowest, about $4.10 \times 10^{-12} \text{ m}^2/\text{s}}$ and $3.4 \times 10^{-12} \text{ m}^2/\text{s}}$ at

28 d and 91 d, respectively. Compared with A1 ordinary tap water concrete, the Cl⁻ penetration coefficients of B1, B2 and B3 AEW concrete reduced 10.5%, 7.8% and 5.3% at 91 d, respectively. The improvement effect of AEW on chloride ion penetration resistance of concrete was relatively obvious, the AEW concrete has a better anti-chloride ion permeability.



Figure 9: Weight loss ratio of concrete



Figure 10: Chloride permeability device



Figure 11: Chloride permeability coefficient of concrete

This is might be because that introducing the electrolyzed water in concrete can promote the cement hydration of concrete and produce the more hydration products to improve the internal pore structure and compactness of concrete, the Cl⁻ penetrability resistance can be improved to a certain extent. Mandal et al. [39] also reported that the durability of the concrete made with electrolyzed water is better due to lower ingress of Cl⁻ ions through the denser microstructure and thicker passive layer. It will be further researched by combining mercury injection and pore distribution test.

3.5 Carbonation Depth of Different Series of Concrete

The carbonation depths of different series of concrete are shown in Fig. 12. It can be seen that the carbonation performance of concrete can be improved by AEW. At the early age of hydration, the carbonation depths of concrete with AEW series were significantly lower than those of concrete with ordinary tap water series at same proportioning. Compared with A1 ordinary tap water concrete, the carbonation depths of B1, B2 and B3 AEW concrete at 3 d were reduced, which were 0.2, 0.6 and 0.8 mm, respectively. In contrast, at the late age of hydration, the difference in carbonation depth between the two series of concrete decreased. The carbonation depths of A1, A2 and A3 concrete at 91 d were 3.2, 3.5 and 4.1 mm, the carbonation depths of B1, B2 and B3 AEW concrete were 2.8, 3.3 and 3.8 mm.



Figure 12: Carbonation depth of different series of concrete

Moreover, with the increase of mineral admixture content, the carbonation depths of two series of concrete both gradually increased, this is probably related to the pore distribution and pore structure of concrete, which can be observed by the SEM micromorphology of concrete cured for 28 d in Fig. 13. From Fig. 13, it can be seen that the pore structure distribution of AEW concrete at each mix proportion was more developed and more average, the porosity of AEW concrete decreased compare with that in natural tap water concrete, this indicated that AEW can promote the hydration reaction to produce more hydration products and make the internal pore structure more dense. Chakraborty et al. [40]. believed that the enhanced durability of electrolyzed water concrete is attributed to the lower ingress of deleterious agents through the less porous and compact microstructure of the hardened composites, which is also consistent with the above result. With the increase of mineral admixture content, the porosity of two series of concrete both gradually increased, which provided a channel for the invasion of carbon dioxide in concrete, resulting in the increase of carbonization depth.



Figure 13: Pore structure distribution of different series of concrete by SEM micromorphology (10000 times)

3.6 X-ray Diffraction Analysis of Different Series of Concrete

To determine the mineral composition of concrete and assess the hydration process more accurately, an X-ray diffraction (XRD) analysis of different cement paste specimens for concrete was conducted in this study. Based on the above results of compressive strength and durability, AEW can obviously improve the early property of concrete, so the 7 d XRD testing for different cement paste specimens was conducted, the XRD patterns are shown in Fig. 14. It can be seen that three kinds of AEW concretes have obvious diffraction peaks of Afm and ettringite, the diffraction peaks of Afm and ettringite in natural water concrete were very weak. Meanwhile, the Ca(OH)₂ diffraction peaks of AEW concretes are enhanced obviously than those of natural water concrete, and the XRD diffraction intensity of hydrated cementitious products such as C₃S and C-A-S-H gels are obviously increased. The diffraction intensity CaCO3 (calcite) in AEW concrete was strong, which can shorten the induction period of C3S and partially participate in the hydration. This result is essentially in agreement with the previous results that AEW may lead to a comparatively more Ca(OH)₂ and C-S-H hydrated products than natural water cement paste [27]. Moreover, the calcium potassium gypsum K₂Ca₅·(SO₄)₆·H₂O and potassium feldspar K₂O·Al₂O₃·SiO₂ were also found easily in the XRD patterns of AEW concrete, indicating that calcium hydroxide in alkaline electrolyzed water react further with hydration products of cementitious materials, which may be beneficial to the growth of early strength of concrete. In addition, the diffraction peak intensity of hydrated products of natural tap water and AEW concrete both gradually decreased with the increase of mineral admixture content.

3.7 SEM Micromorphology Analysis of Different Series of Concrete

The SEM micromorphology of different series of concrete at 7 d is observed in Fig. 15. It can be seen that the microstructure of A1 concrete without mineral admixtures basically presented to be flaky and tabular, but the micromorphology of B1 AEW concrete was obvious cluster and granulation instead. This may be related to the charge adsorption of AEW on the cement surface, the electron layer adsorbed on the surface of cement particles can make the cement particles evenly disperse due to electrostatic repulsion [41]. Moreover, based on the results of XRD analysis, the alkaline electrolyzed water can promote the cement hydration and stimulate the activity of mineral admixture, probably to produce more C-A-S-H, C-S-H

gels, $Ca(OH)_2$ and other hydration products, the $Ca(OH)_2$ and AFt phases were more easily observed in the micromorphology of AEW cement pastes. The flocculent C-S-H gel intercalated with the needle AFt phase, which made the structure of the cement paste denser and benefit the increase of strength [42]. The microstructure of B2 and B3 AEW concrete with mineral admixtures was more developed and more average than A2 and A3 concrete. The result clearly justifies the appearance of the microstructural compactness of AEW cement paste.



Figure 14: 7 d X-ray diffraction patterns of different cement paste specimens



Figure 15: SEM images of different series of concrete (10000 times)

4 Conclusions

Through a comprehensive analysis, the following conclusions can be drawn in this study:

- (1) Alkaline electrolyzed water (AEW) can accelerate hydration process of cement in paste, the cement particles are absorbed and dispersed to release more free water, leading to a workability improvement of AEW concrete than that of natural tap water concrete.
- (2) AEW in concrete can accelerate cement hydration and activate the pozzolanic effect of mineral admixtures, leading to an increase of compressive strength at early age. Compared with natural tap water cement concrete, when the replacement ratio of double-doped mineral admixture to cement are 0%, 40% and 50%, the strength of alkaline electrolyzed water concrete increased by 13.6%, 7.3% and 2.6% at 3 d, only 4.2%, 2.5% and 1.1% at 28 d.
- (3) Introducing the AEW can also improve the internal pore structure and compactness of concrete, the anti-chloride permeability, frost resistance, carbonization resistance and durability of alkaline electrolyzed water concrete with high volume mineral admixture are all lower than those of natural tap water cement concrete.
- (4) The XRD and SEM results indicate that AEW can promote the hydration reaction of cement to represent comparatively more hydrated products such as C-S-H gels, Ca(OH)₂ and ettringite. At the same time, the calcium potassium gypsum K₂Ca₅·(SO₄)₆·H₂O and potassium feldspar K₂O·Al₂O₃·SiO₂ are also observed easily at the early age, which can benefit the improvement of strength and durability.
- (5) The fundamental mechanism of AEW to activate the pozzonlanic effect of mineral admixtures still needs further study. From the economic and environmental benefit, the AEW probably could realize the effective utilization of about 50% mineral admixture amount of concrete without strength loss, leading to reduce the cost for cement production and associated CO₂ emission, which has broad application prospect.

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