

Effect of shrinkage reducing admixture on drying shrinkage and durability of alkali-activated coal gangue-slag material

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Abstract: This paper studied the effect of shrinkage reducing admixture (SRA) on drying shrinkage and microstructure of alkali-activated coal gangue-slag (AACGS). Concrete was prepared by using calcined coal gangue as coarse aggregate. The effect of SRA on the compressive strength and durability of concrete under different calcined coal gangue coarse aggregate (CCGCA) content was investigated, and the action mechanism of SRA was also systematically analyzed. The research results showed that the addition of 3% SRA can reduce the drying shrinkage of AACGS mortar by 47.5%. The 28d compressive strength of concrete can be reduced by mixing with SRA, which has limited effect on the 90d compressive strength. SRA can significantly improve the frost resistance durability of concrete (water freezing and salt freezing conditions can increase the number of freeze-thaw cycles by about 50 and 25, respectively) and sulfate attack resistance (90d testing). With the increasing content of CCGCA, the effect of adding SRA on the improvement of anti-chloride penetration has become more obvious. SRA caused changes in the pore size distribution, reaction

rate and interfacial bond strength of AACGS concrete. Moreover, the present study also provides the experimental and detailed theoretical basis for the action mechanism of SRA in alkali-activated materials concrete.

Key words: shrinkage reducing admixture; alkali-activated coal gangue-slag; calcined coal gangue coarse aggregate; compressive strength; durability; action mechanism

1 Introduction

Alkali-activated materials have the advantages of low hydration heat, low environmental impact, early high strength, chemical attack resistance, and good durability, etc., which have become the most promising construction materials to replace Portland cement [1-3]. Due to the reaction mechanism of alkali-activated materials is different from that of ordinary Portland cement, its stability is not as good as that of cement materials, posing some challenges to the popularization and application of alkali-activated materials. The shrinkage characteristics of concrete will contribute to a decrease of compressive strength and reduce the durability of concrete. It has been proved that the shrinkage of alkali-activated materials is significantly higher than that of cement-based materials [4-6], which seriously hinders the promotion and application of alkali-activated materials in concrete.

Coal gangue is regarded as a kind of hard black-gray rock associated with coal seam, which is a solid waste discharged during coal mining and coal washing. Its main chemical components are SiO_2 and Al_2O_3 [7]. Coal gangue is restricted by the geographical location, environment and production benefits, and natural accumulation is the most extensive treatment method, which not only occupies land but also pollutes water, air, and soil, adversely affecting the environment [8,9].

At present, coal gangue is mainly used for production of cement [10], road construction [11] and

sintered bricks [12]. Thus, its utilization rate is still very low. To solve the problem of idleness and pollution of coal gangue in countries producing coal, it is urgent to find a new way to improve the utilization rate of coal gangue. The volcanic ash activity of coal gangue requires high-temperature calcination to stimulate, different from fly ash and slag, coal gangue contains crystallized state and non-crystallized state, and the crystallized state plays a filling role in the pastes. Zhou et al. [13] used spontaneous combustion coal gangue coarse aggregate instead of ordinary crushed stone to prepare concrete, which could satisfy the requirements of compressive strength grade, finding that the interface transition zone (ITZ) between spontaneous combustion coal gangue and cement mortar were well bonded. Guan et al. [14] prepared concrete by using coal gangue as coarse aggregate, and found that with the increase of the content of coal gangue coarse aggregate, the frost resistance and mechanical properties of concrete decreased, and the toughness, damage layer thickness and peak stress-strain of concrete all decreased. Zhang et al. [15] prepared concrete by using spontaneous combustion coal gangue as coarse aggregate, and found that the coal gangue coarse aggregate size has limited influence on concrete. When the content of coal gangue coarse aggregate is 100%, the compressive strength, splitting tensile strength and elastic modulus of concrete decrease by 19.4%, 36.1% and 32.2% respectively. The application of coal gangue in concrete not only has good economic benefits, but also protects the environment, of which the environmental benefits are far greater than its economic benefits. Therefore, it is of great significance to expand the application of coal gangue in concrete.

The research group has studied the drying shrinkage of alkali-activated coal gangue-slag (AACGS) mortar, finding that when the modulus of alkali activator is 1.3 and the mixing ratio of coal gangue and slag is 1:1, the drying shrinkage rate of AACGS mortar is 3.05 times that of cement

mortar [16]. Such a high drying shrinkage rate greatly limits the engineering application of AACGS materials. Till now, the research and measures to reduce drying shrinkage of alkali-activated materials have been greatly limited, including expansion compensation, reduction of capillary pressure and fiber skeleton inhibiting shrinkage, etc [17-20]. Hu et al. [21] found that shrinkage reducing admixture (SRA) significantly reduced the autogenous shrinkage and drying shrinkage of alkali-activated slag mortar, and decreased the capillary tension of mortar specimens. Palacios et al. [22] found that in the relative humidity of 99% and 50%, SRA can reduce the shrinkage of alkali-activated slag mortar by 85% and 50% respectively, indicating that it could be clearly seen that SRA had more significant shrinkage reduction performance under high curing humidity conditions. That is to say, the SRA can not only reduce drying shrinkage, but also significantly improve chemical shrinkage or autogenous shrinkage under high humidity. Bentz et al. [23] investigated the effect of SRA on the internal relative humidity of cement-based mortar, concluding that in the low water-cement ratio mortar cured under sealed conditions, the addition of SRA led to the reduction of internal relative humidity and the significant reduction of autogenous shrinkage. Darshan et al. [24] found that the SRA can increase the internal relative humidity in addition to reducing the surface tension of the pore solution, and this is very effective in reducing the autogenous shrinkage of alkali-activated slag mortar. Compared with ordinary crushed stone, coal gangue exhibits superior water absorption performance as concrete coarse aggregate [25]. Song et al. [26] studied the effects of superabsorbent polymers on the internal relative humidity and autogenous shrinkage of alkali-activated slag mortar, finding that increasing the content of superabsorbent polymers in mortar is conducive to improving the internal relative humidity of mortar and reducing shrinkage. Vlastimil et al. [27] found that SRA has certain incompatibility with alkali-activated slag materials, and SRA

1 89 reduces drying shrinkage by delaying hydration reaction, leading to pore structure coarsening.

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3 90 The addition of SRA will cause a series of changes in the mortar, including pore size
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6 91 distribution, reaction rate, interfacial bond strength, etc. At present, SRA has positive effects on
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9 92 chloride permeability and frost resistance of cement concrete [28,29]. In the study on alkali-
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12 93 activated materials, SRA focuses on the hydration and drying shrinkage of alkali-activated pastes or
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15 94 mortar [30]. A lot of researchers have studied the action mechanism of SRA to reduce capillary
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18 95 tensile stress through reducing the surface tension of pore water [31-33]. However, there is still a
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21 96 lack of relevant experimental and theoretical research about whether it changes pore distribution
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24 97 and what impact it has on the durability of alkali-activated concrete. Theoretically, although SRA
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27 98 inhibition of the shrinkage of alkali-activated materials will improve the durability of concrete, the
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33 100 SRA to restrain dry shrinkage of AACGS mortar still remains blank. Therefore, even though SRA
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36 101 has a positive effect on alkali-activated materials, coal gangue materials are different from other
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39 102 research materials. Therefore, it is necessary to deeply explore the effect and action mechanism of
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42 103 SRA on AACGS mortar and concrete.

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44 104 In the current work, high-calcium slag and silicon-aluminum coal gangue materials were used
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47 105 as cementitious materials. Sodium hydroxide and sodium silicate were used as alkali activators, and
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50 106 calcined coal gangue at 700°C was used as coarse aggregate to prepare AACGS mortar and concrete
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53 107 respectively. The effect of SRA dosage (1%, 2% and 3%) and the content of calcined coal gangue
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56 108 coarse aggregate (CCGCA) (0%, 30%, 50%, 70% and 100%) on the drying shrinkage characteristics
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59 109 of AACGS mortar and the durability of concrete were explored. The durability tests included rapid
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62 110 chloride migration coefficient method (RCM), freeze-thaw resistance (water-freeze and salt-freeze)

111 and sulfate attack resistance tests. Using the mercury intrusion porosimetry (MIP) method and
112 scanning electron microscope (SEM), the effect of SRA on the microstructure of AACGS was
113 deeply investigated, and the mechanism of SRA to improve the durability of AACGS concrete was
114 deeply analyzed.

115 **2 Experimental**

116 **2.1 Raw materials**

117 The coal gangue powder and coarse aggregate were calcined in muffle at 700°C for two hours,
118 Fig.1 shows the photos of coal gangue coarse aggregate used in the test. Fig.1(a) shows the raw coal
119 gangue, which is black-gray. Fig.1(b) shows the calcined coal gangue at 700°C, which is gray-white.
120 The S95 granulated blast furnace slag from HBIS Group Co., Ltd. was used. The chemical
121 compositions of the two materials determined by X-ray fluorescence (XRF) are shown in Table 1.
122 The particle size distribution of the two materials were determined by laser particle size distribution
123 instrument, as shown in Fig.2. The median particle size d50 of coal gangue powder and slag powder
124 were 17.252μm and 10.529μm, respectively. SRA uses alkyl polyether SRA (liquid solvent, the main
125 components include aliphatic ethylene diether and hydroxide alkylamines), which is provided by
126 China Fuyuan Construction Material Factory. The 96% sodium hydroxide analytical pure (NH) and
127 sodium water glass (NS, 26.5%SiO₂, 8.5%Na₂O, 65%H₂O, modulus 3.22) were selected as alkali
128 activator. The chemicals reagent used in the durability test included 99.5% sodium chloride solution
129 (NaCl) analytical pure and 99.9% Na₂SO₄ analytical pure.



(a) Raw coal gangue (b) Calcined coal gangue

Fig.1 Photos of CCGCA for test

Table 1 The main composition of coal gangue and slag /wt%

Parameters	Coal gangue	Slag
SiO ₂	56.56	30.58
Al ₂ O ₃	36.78	14.04
CaO	0.62	38.43
Fe ₂ O ₃	1.95	0.35
MgO	0.22	10.57
Na ₂ O	0.42	0.57
SO ₃	0.03	2.36
TiO ₂	2.10	1.93
LOI	1.32	1.17

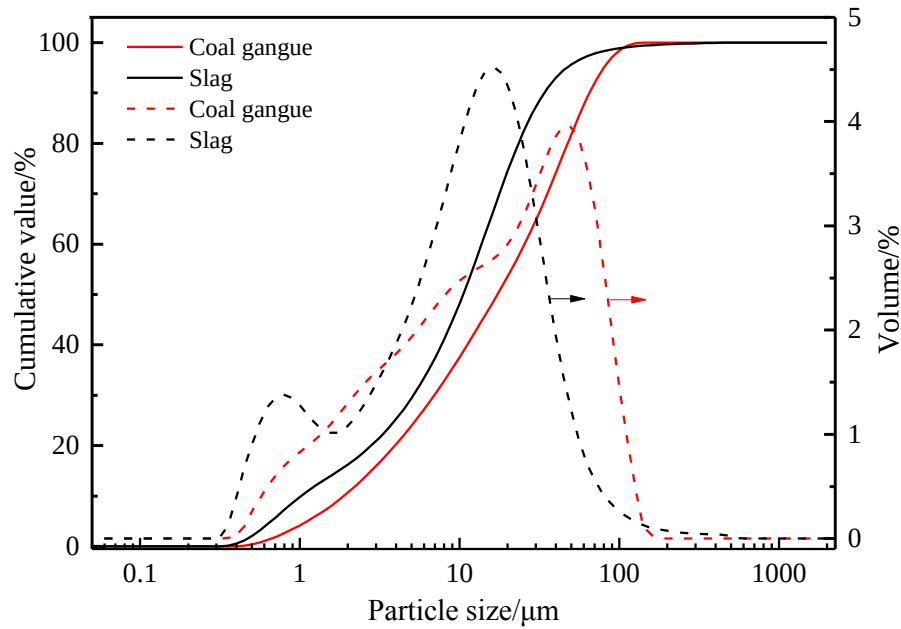


Fig.2 Coal gangue and slag powder particle size analysis

2.2 Mixture proportions and specimens preparation

In this paper, the preparation of specimens is divided into two types: AACGS mortar and

1 138 AACGS concrete. Mortar specimens are used for the determination of drying shrinkage, while
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3 139 concrete specimens are used for the determination of compressive strength and durability. The alkali
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6 140 activator modulus of all specimens was 1.30, and the mixing proportion of coal gangue to slag
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9 141 powder were 1:1. A total of 4 groups of mortar specimens and 12 groups of concrete specimens
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11 142 were prepared, and the specific mixing proportion was shown in Table 2 and Table 3. The alkali-
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14 143 activated solution was prepared 24 hours before use, and cooled to room temperature to ensure that
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17 144 the activator components were mixed evenly and the polymerization process was improved [34].
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20 145 The required alkali activator modulus was obtained by mixing sodium hydroxide analytical pure
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23 146 and sodium silicate solutions, and adding additional water to prepare the product conforms to the
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26 147 liquid-solid ratio requirements in Table 3.

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28 148 The preparation of the specimens all adopts the forced mixing method. The coal gangue and
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31 149 slag powder, standard sand (fineness modulus 2.70) were mixed and stirred for 3min, then pour into
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34 150 alkali activator solution and continue to stir for 5min. The newly mixed mortar was poured into the
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37 151 test mold with embedded metal nails at both ends to test the drying shrinkage values of different
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39 152 ages. Mix coal gangue and slag powder, standard sand, ordinary crushed stone and CCGCA for
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42 153 5min, then pour into the alkali activator solution and continue to stir for 5min. Pour fresh AACGS
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45 154 concrete into the corresponding test mold, and the dimension and quantity of each group of
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48 155 specimens are shown in Table 4. All specimens were cured in the standard curing room ($RH=95\pm1\%$,
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51 156 $T=20\pm2^{\circ}C$) for 1d and then demoulded. The curing conditions of subsequent specimens were divided
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56 158 a: After demolding the mortar specimen, the initial length L_0 and the initial mass W_0 were
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59 159 determined by the micrometer specific length meter (accuracy 0.001mm) and the electronic scale
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(accuracy to 0.01g). The specimens were moved to the cement mortar drying shrinkage curing humidity control box with a temperature of $20\pm 2^{\circ}\text{C}$ and relative humidity of $60\pm 5^{\circ}\text{C}$, and the length values of 1d-180d L_t and mass W_t were tested respectively.

b: Palacios et al. [22] found that SRA reduced shrinkage performance more significant under high curing humidity conditions. Therefore, AACGS concrete specimens would continue to be moved to the standard curing room for curing until the measured age.

Table 2 Mix proportion of AACGS mortar specimens

No.	Coal gangue	Slag	Standard sand	NH	NS	H ₂ O	SRA
SRA-0	500	500	2750	94.17	580.85	188.35	—
SRA-1	500	500	2750	94.17	580.85	188.35	1%
SRA-2	500	500	2750	94.17	580.85	188.35	2%
SRA-3	500	500	2750	94.17	580.85	188.35	3%

Table 3 Mix proportion of AACGS concrete specimens

No.	Coal	Slag	Standard	Coarse aggregate		Liquid	Alkali-	Modulus	SRA
	gangue	powder	sand	Crushed	Coal	-solid	activator		
	powder			stone	gangue	ratio	solid dosage		
C-SRA-0	280	280	565	1050	—	0.40	0.32	1.30	—
C-SRA-1	280	280	565	1050	—	0.40	0.32	1.30	1%
C-SRA-2	280	280	565	1050	—	0.40	0.32	1.30	2%
C-SRA-3	280	280	565	1050	—	0.40	0.32	1.30	3%
CG30	280	280	565	735	315	0.40	0.32	1.30	—
CG50	280	280	565	525	525	0.40	0.32	1.30	—
CG70	280	280	565	315	735	0.40	0.32	1.30	—
CG100	280	280	565	—	1050	0.40	0.32	1.30	—
CG30-SRA	280	280	565	735	315	0.40	0.32	1.30	3%
CG50-SRA	280	280	565	525	525	0.40	0.32	1.30	3%
CG70-SRA	280	280	565	315	735	0.40	0.32	1.30	3%
CG100-SRA	280	280	565	—	1050	0.40	0.32	1.30	3%

Table 4 The dimension and quantity of specimens prepared for each group

Experiment type		Dimension		Quantity	Total
AACGS mortar	Drying shrinkage	25mm×25mm×280mm		4	16
AACGS	Compressive strength	100mm×100mm×100mm		6	72
	Rapid chloride	Φ100mm×50mm		3	12

concrete	penetration			
	Freeze-thaw cycles	100mm×100mm×400mm	3	36
	(water)			
	Freeze-thaw cycles	100mm×100mm×400mm	3	36
	(salt)			
	Sulfate attack	100mm×100mm×100mm	6	72
Total				244

2.3 Test methods

I: Drying shrinkage

According to the initial length of the tested specimens L_0 and the length of each curing age L_t , the initial mass W_0 and the mass of each curing age W_t , the drying shrinkage ratio ε_s and mass loss ΔW are calculated as shown in Equations 1 and 2:

$$\varepsilon_s = (L_0 - L_t) / L_0 \times 100\% \quad (1)$$

$$\Delta W = (W_0 - W_t) / W_0 \times 100\% \quad (2)$$

The arithmetic mean value of the three specimens is taken as the drying shrinkage value. When the deviation between the test value and the mean value is greater than 20%, the value should be removed.

II: Compressive strength

According to GB/T 50107-2010 standard for test methods, the compressive strength test was conducted by YAW-300 pressure tester, and the specimens with curing ages of 28d and 90d were tested respectively. The loading speed is 0.5MPa/s, three specimens were tested in per group, and the compressive strength is the mean value of the three specimens.

III: Rapid chloride penetration

According to GB/T 50082-2009 standard for test methods, the RCM method was used to test the chloride ion diffusion coefficient of each specimen, and the test process was conducted in accordance with GB/T 50082-2009. Rapid chloride ion penetration test specimen size of $\Phi 100$

mm×50 mm (three Φ100mm×50mm specimens are cut out from the middle of Φ100mm×200mm),

chloride ion diffusion coefficient D calculation is shown in Equation 3:

$$D = \frac{0.0239 \times (273 + T)L}{(U-2)t} (X_d - 0.0238 \sqrt{\frac{(273 + T)LX_d}{U-2}}) \quad (3)$$

Where: U—absolute value of the voltage used /V;

T—Mean value of initial temperature and end temperature of anode solution /°C;

X_d—Mean value of chloride ion penetration depth. Ten penetration depths were measured at equal distances and accurate to 0.1mm;

t—Test duration /h.

IV: Freeze-thaw cycles

In order to explore the effect of SRA on the durability of AACGS concrete, the freeze-thaw test was conducted by water freezing and salt freezing, and 5% NaCl was used in the salt freezing test. According to GB/T 50082-2009 standard for test methods, After the specimens under standard curing for 24d were taken out, then immersed in water for 4d. The freeze-thaw cycles were carried out by water freezing and salt freezing respectively. The mass and relative dynamic elastic modulus of the specimens were tested per 25 freeze-thaw cycles. Take the relative dynamic elastic modulus loss rate and mass loss rate as the evaluation criteria, the relative dynamic elastic modulus ΔP and mass loss rate Δw of a single specimen is calculated as shown in Equations 4 and 5:

$$\Delta P = (f_n^2 - f_0^2) / f_0^2 \times 100\% \quad (4)$$

$$\Delta w = (w_0 - w_n) / w_0 \times 100\% \quad (5)$$

Where: f₀—The initial transverse fundamental frequency of concrete specimen before freeze-thaw cycles test /Hz;

f_n—The transverse fundamental frequency of concrete specimen after n freeze-thaw cycles

/Hz;

w_0 —Mass of concrete test block before freeze-thaw cycles test/g;

w_n —Mass of concrete test block after n freeze-thaw cycles test/g;

V: Sulfate attack

According to GB/T 50082-2009 standard for test methods, 5% Na₂SO₄ solution was used for sulfate attack, and AACGS concrete with curing age of 28d was fully immersed in Na₂SO₄ solution for 90d, and specimens of the same batch were immersed in water for 90 days. The compressive strength of AACGS concrete after sulfate attack and water immersion for 90 days was tested, and the compressive strength loss rate was taken as the test result of sulfate attack [35]. Compressive strength loss rate ΔC is calculated as Equation 6:

$$\Delta C = (C_w - C_s) / C_w \times 100\% \quad (6)$$

Where: C_s —Compressive strength after 90 days of sulfate attack /MPa;

C_w —Compressive strength after water immersion for 90 days /MPa.

VI MIP testing

Specimens for curing for 28 days were broken and cored by hammering method, and about 25 pieces of 1cm-2cm blocks were taken from per group. Then isopropanol was used to terminate hydration and low-temperature vacuum drying was performed for 24h. Using AutoPore IV automatic mercury porosimeter, continuous mercury entry (contact angle and surface tensions are 130° and 485 dynes/cm, respectively). The pore diameter distribution curve of mortar specimens was obtained by changing the injection pressure.

VI SEM testing

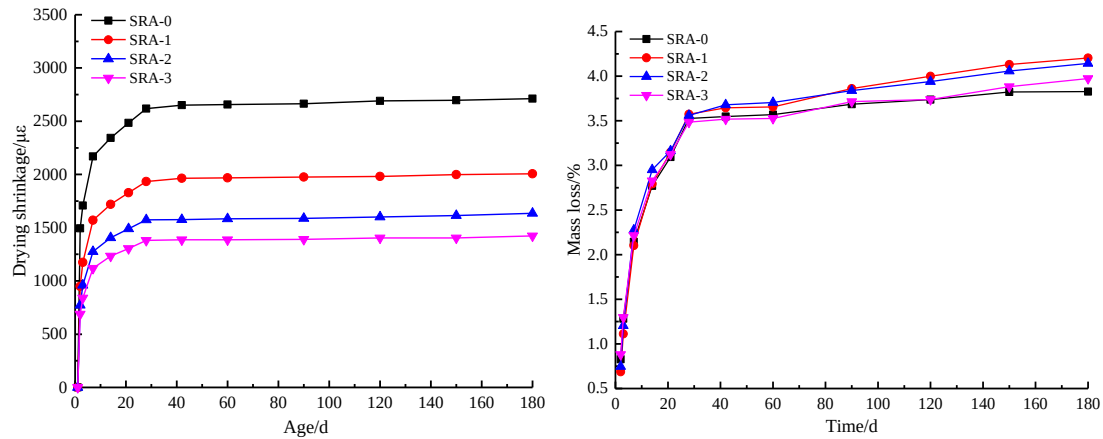
In order to explore the effect of SRA on the internal structure of AACGS concrete and the

bonding interface between AACGS mortar and CCGCA, the core (bonding interface between AACGS mortar and CCGCA, sampling contains both mortar and part of the coal gangue particles) was taken from the concrete after curing for 28 days (about 1-2cm), and the hydration was stopped by using isopropanol, and then dried in a low-temperature vacuum for 24 hours. The field emission scanning electron microscope of Hitachi SU8010 was used for observation. The specimen surface was sputtered with gold-plated palladium before imaging, and then the surface morphology of the broken specimen was observed by SEM.

3 Test results analysis

3.1 Drying shrinkage of AACGS mortar

Fig.3 shows the drying shrinkage test results of AACGS mortar, and Fig.3(a) is the drying shrinkage value. Compared with SRA-0 specimen, when SRA dosage is 1%, 2% and 3%, the drying shrinkage of AACGS mortar decreases by 26.01%, 39.66% and 47.50%, respectively. Obviously, the drying shrinkage of AACGS mortar decreases with the increase of SRA dosage. When the SRA dosage was 3%, the drying shrinkage decreased by 47.50%, which was similar to the experimental results of Palacios et al [22, 30]. Fig.3(b) is the result of mass loss, showing that at the initial stage of drying shrinkage of AACGS mortar specimens, the mass loss rate of specimens increases with SRA dosage, and the SRA dosage has no significant effect on the mass loss rate. Furthermore, Comparing Fig.3 (a) and (b), there is no significant relationship between drying shrinkage and mass loss rate. The effect of SRA on inhibiting drying shrinkage of AACGS mortar is not directly related to water loss.



(a) Drying shrinkage (b) Mass loss
Fig.3 Effect of SRA on drying shrinkage and mass of AACGS mortar

3.2 Compressive strength analysis

The compressive strength of concrete is associated with the strength of the mortar matrix, the coarse aggregate and the bonding strength of mortar matrix-coarse aggregate [36,37]. Table 5 shows the compressive strength results of AACGS concrete. The compressive strength decreases with the increase of SRA dosage. Compared with the C-SRA-0 specimen, the 28d compressive strength of AACGS concrete decreased by 3.38%, 4.49% and 6.73% when SRA were added 1%, 2% and 3%, respectively, mainly due to SRA hindered the early polymerization process of AACGS materials. It was found that the compressive strength of 90d of each AACGS concrete specimens was differed by less than 3%, which indicated that SRA had little effect on the 90d compressive strength of concrete.

Furthermore, the compressive strength of AACGS concrete decreases with the increase of the content of CCGCA. When the content of CCGCA is 30%-50%, the compressive strength of mixed SRA concrete specimens is greater than that of unmixed SRA concrete specimens. Under the same content of CCGCA, the difference between specimens is only reflected in the bond strength between the AACGS mortar matrix-CCGCA, indicating that the addition of SRA can enhance the bond strength between AACGS mortar matrix-CCGCA. The compressive strength of 90d showed that

the AACGS concrete mixed with SRA specimens were stronger than that of 28d. In general, the addition of SRA reduced the short-term compressive strength of AACGS concrete, while it had little effect on the 90d compressive strength of AACGS concrete. Moreover, the addition of SRA enhanced the bond strength of AACGS mortar matrix-CCGCA.

Table 5 The compressive strength of AACGS concrete

No.	Compressive strength		No.	Compressive strength	
	28d/MPa	90d/MPa		28d/MPa	90d/MPa
C-SRA-0	62.12	66.80	CG70	55.10	55.30
C-SRA-1	60.02	65.99	CG100	49.33	48.70
C-SRA-2	59.33	65.42	CG30-SRA	58.08	60.06
C-SRA-3	57.94	64.81	CG50-SRA	57.76	58.78
CG30	58.37	59.28	CG70-SRA	56.37	56.49
CG50	57.65	57.83	CG100-SRA	51.27	51.44

3.3 Rapid chloride penetration analysis

Fig.4 shows the chloride ion diffusion coefficient of AACGS concrete, and Fig.4(a) is the SRA dosage factor. Obviously, the chloride ion diffusion coefficient of AACGS concrete with 3% SRA dosage is the highest without the addition of CCGCA, and the addition of SRA generally increases the diffusion coefficient of chloride ions. However, the chloride ion diffusion coefficient of the four specimens were around $3.05 \times 10^{-12} \text{m}^2 \text{s}^{-1}$, showing no significant difference. Fig.4(b) shows the factors of CCGCA content. When the content of CCGCA is 0%, 30%, 50%, 70% and 100%, the chlorine ion diffusion coefficient decreases by -1.45%, -0.39%, 0.71%, 6.19% and 21.55% respectively after the addition of SRA. When the content of CCGCA is more than 50%, the improvement effect of adding SRA becomes more obvious. The main reason is that the active ingredients SiO_2 and Al_2O_3 in calcined coal gangue are the same as that in calcined coal gangue powder, which also participates in the alkali-activated reaction process, thus enhancing the interface bite force between AACGS mortar and CCGCA. The addition of SRA reduced the shrinkage of AACGS mortar, which effectively improved the pore structure of the bonding interface of AACGS

mortar-CCGCA.

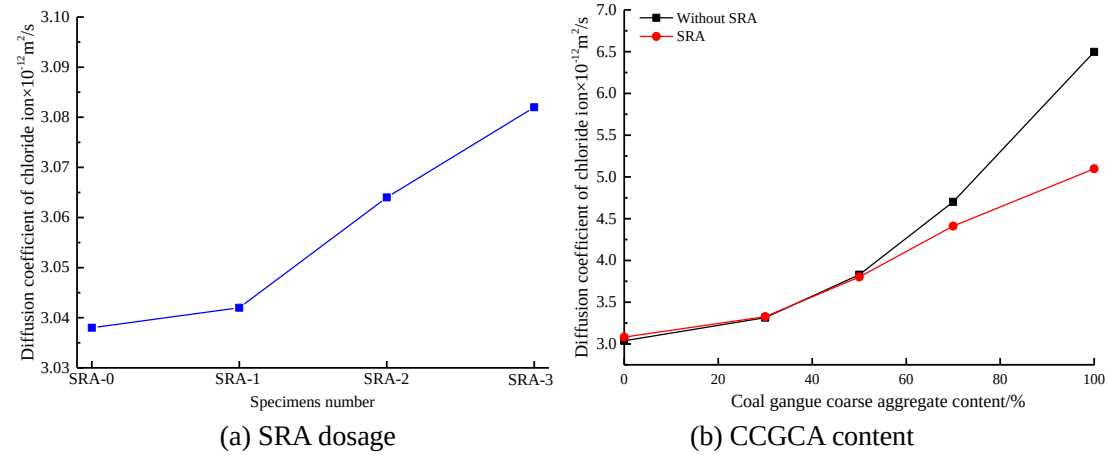


Fig.4 Chlorine ion diffusion coefficient of AACGS concrete

3.4 Freeze-thaw cycles analysis

3.4.1 Water freeze-thaw cycles

Frost resistance test is an important index to evaluate concrete durability [38]. Fig.5 shows the water freeze-thaw cycles test results of AACGS concrete with different SRA dosages. It's obvious that the dynamic elastic modulus of AACGS concrete unmixed SRA is lower than 60% after 200 freeze-thaw cycles, and the frost resistance grade is less than F200. When mixed with SRA, AACGS concrete has more than 200 freeze-thaw cycles, and the frost resistance grade is over F200. Furthermore, the frost resistance durability of AACGS concrete was enhanced with the increasing SRA dosage. Fig.5(b) shows that mass loss rate gradually increases when the freeze-thaw cycles increase. Additionally, the mass loss rate of AACGS concrete decreased with the increase of SRA dosage. In comparison with C-SRA-0 specimen, the mass loss rate of 1%, 2% and 3% of SRA dosage decreased by 18.90%, 27.05% and 34.54% respectively after 200 freeze-thaw cycles.

Fig.6 shows the results of water freeze-thaw cycles test of AACGS concrete under different CCGCA contents. Fig.6(a) is the results of relative dynamic elastic modulus. Under the condition of the same proportion of CCGCA, AACGS concrete mixed with SRA has better frost resistance.

When the content of CCGCA is 30%, AACGS concrete unmixed SRA will be damaged after 150 freeze-thaw cycles. However, the freeze-thaw cycle can exceed 175 cycles after the addition of SRA. When the content of CCGCA exceeds 30%, compared with AACGS concrete unmixed SRA, the freeze-thaw cycle of AACGS concrete mixed with SRA increased by at least 50 cycles. Fig.6(b) shows the mass loss rate results. The test results show the same variation rule as the relative dynamic elastic modulus loss. Under the condition of the same freeze-thaw cycles and the content of CCGCA, the mass loss rate of AACGS concrete mixed with SRA is lower.

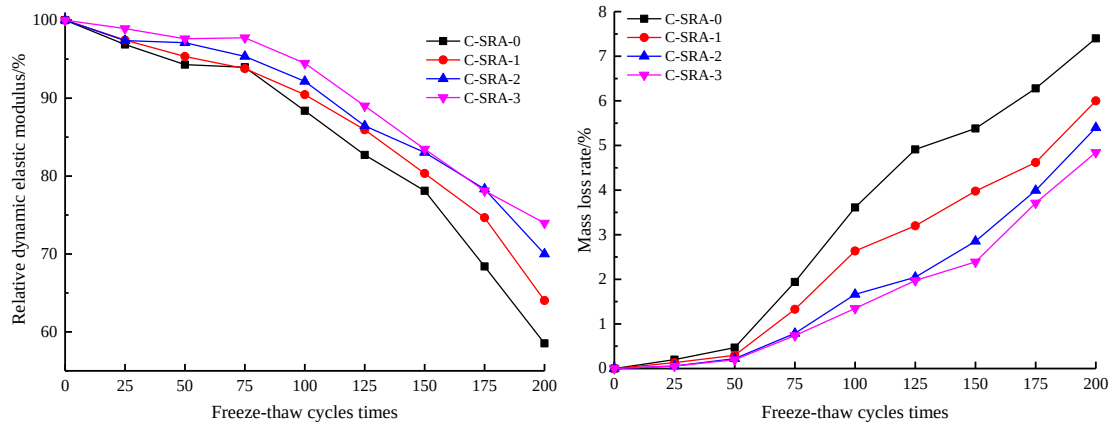


Fig.5 Results of water freeze-thaw cycles test of AACGS concrete with different SRA dosages

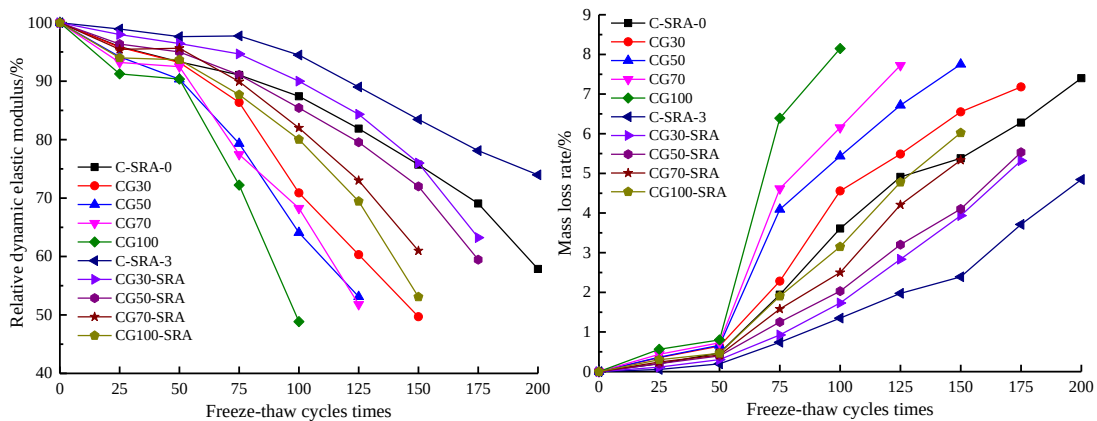


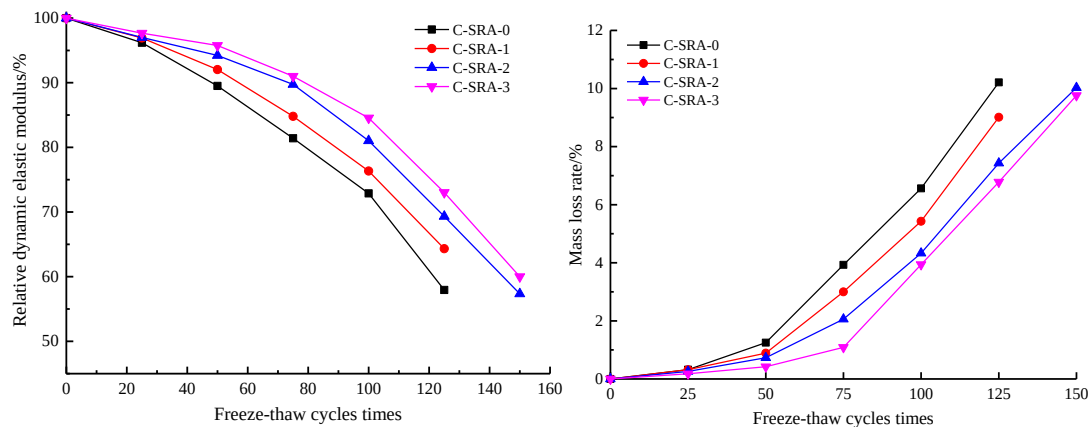
Fig.6 Results of water freeze-thaw cycles test of AACGS concrete with different CCGCA contents

3.4.2 Salt freeze-thaw cycles

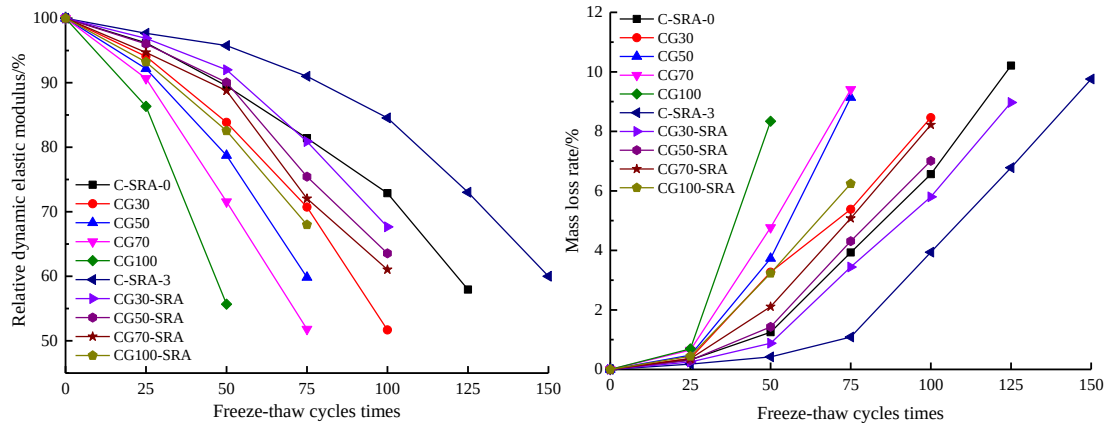
In the actual environment, concrete is often damaged by the combined action of freeze-thaw

cycles and salt erosion. Salt freeze-thaw is more common than water freeze-thaw, and salt freeze-thaw is the main factor which can cause the destruction of concrete [39]. Fig.7 shows the results of salt freeze-thaw cycles test AACGS concrete under different SRA dosages, and the overall performance is the same as that of water freeze-thaw cycles. Under the action of salt freeze-thaw, AACGS concrete has fewer freeze-thaw cycles and lower frost resistance grade. AACGS concrete unmixed SRA has a frost resistance grade of F125, while AACGS concrete mixed with SRA has a frost resistance grade of more than F125. Moreover, even AACGS concrete mixed with 3% SRA has a frost resistance grade of more than F150. In comparison with the mass loss rate under water freeze-thaw cycles, the mass loss rate under salt freeze-thaw cycles is greater.

Fig.8 shows the results of salt freeze-thaw cycles of AACGS concrete under different CCGCA contents, all of which demonstrate that the anti-salt freeze-thaw durability of AACGS concrete becomes worse with the content of CCGCA increases. The effect law of SRA on AACGS concrete is the same as that under water freeze-thaw conditions. Under the condition of salt freeze-thaw cycles, AACGS concrete mixed with SRA obtains 25 more freeze-thaw cycles than that of AACGS concrete unmixed SRA, revealing excellent anti-salt freeze-thaw durability. Fig.8(b) shows the result of mass loss rate. Under the same content of CCGCA and salt freeze-thaw cycles, the mass loss rate of AACGS concrete mixed with SRA becomes lower.



(a) Relative dynamic elastic modulus (b) Mass loss rate
Fig.7 Results of salt freeze-thaw cycles test of AACGS concrete with different SRA dosages



(a) Relative dynamic elastic modulus (b) Mass loss rate
Fig.8 Results of salt freeze-thaw cycles test of AACGS concrete with different CCGCA contents

3.5 Sulfate attack analysis

Ismail et al. [40] attacked the alkali-activated slag-fly ash binary polymer in 5% Na_2SO_4 solution for 90 days, finding that alkali-activated slag-fly ash had almost no damage, and the structure of polymerization product C-A-S-H of the alkali-activated slag-fly ash exposed to Na_2SO_4 was basically intact. Tang et al. [41] found that replacing Portland cement with coal gangue has a positive effect on the sulfate resistance of cement. Fig.9 shows the results of the sulfate resistance test of AACGS concrete, and Fig.9(a) shows the results of the SRA dosages. The compressive strength of AACGS concrete decreases after being attack in 5% Na_2SO_4 solution for 90d when unmixed SRA. After the addition of SRA, the compressive strength of AACGS concrete increased after being attack in 5% Na_2SO_4 solution for 90d. In addition, the higher the SRA dosage, the greater the increase of compressive strength. The main reason is that the addition of SRA slows down the early polymerization process of AACGS concrete, and the larger the SRA dosage, the stronger the inhibition effect, causing much unreacted coal gangue and slag particles are encased in the precipitated cementitious phase. After soaking AACGS concrete in 5% Na_2SO_4 solution, Na_2SO_4 solution can be used as a salt activator to continue to promote the polymerization of AACGS pastes

[42], contributing to an increase in the compressive strength of AACGS after the attack of Na_2SO_4 solution.

Fig.9(b) shows the results of CCGCA content. It can be clearly seen that the addition of CCGCA is beneficial to the sulfate resistance of AACGS concrete, and the sulfate resistance of the same content CCGCA is stronger after the addition of SRA. The AACGS concrete specimen mixed with SRA is strengthened with increasing content of CCGCA, which is mainly due to the subsequent reaction of salt excited AACGS enhances the bond strength of AACGS mortar-CCGCA. Furthermore, without the addition of SRA, after 90 days of sulfate attack, the compressive strength loss rate tends to be stable when the content of CCGCA is greater than 50%. When mixed 3% SRA, the large content of CCGCA still shows an advantage. However, the advantages become smaller.

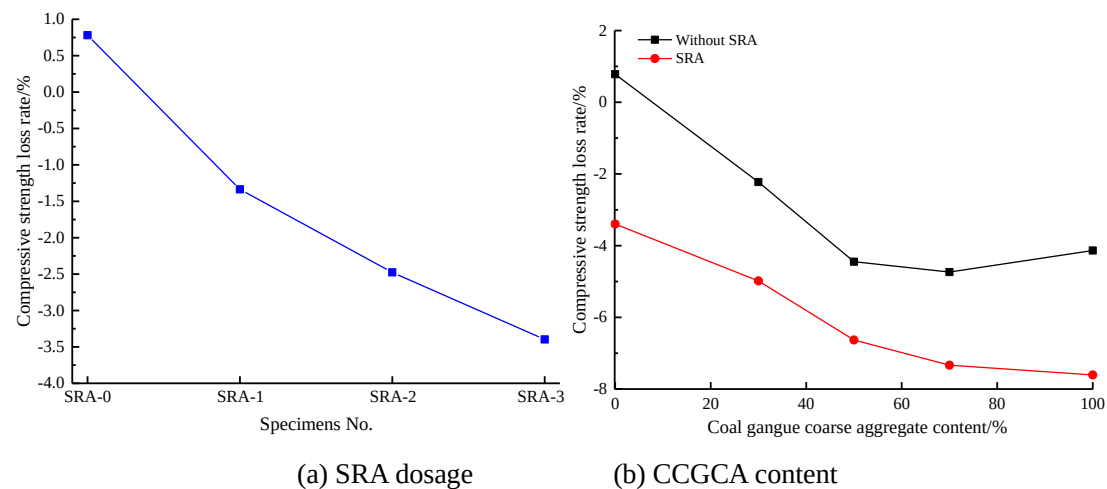


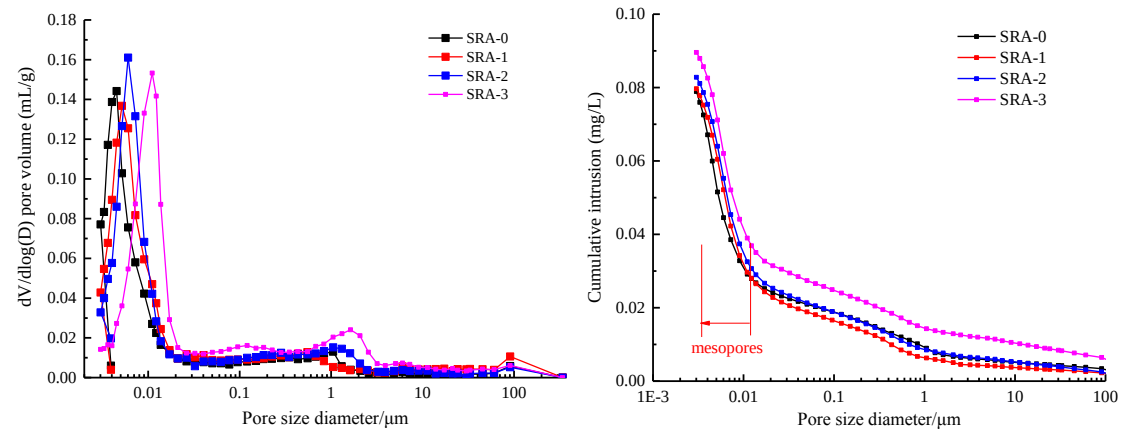
Fig.9 Results of sulfate attack test of AACGS concrete

3.6 MIP analysis

Previously, our research group found that the main reasons for the large drying shrinkage of AACGS mortar were the filling of unreacted coal gangue and alkali solids. The collapse and recombination of the gel resulted in the refinement of the pore size structure of mortar specimens, which resulted in greater capillary tensile stress [16]. Fig.10 shows the MIP test results of AACGS mortar specimens with different SRA dosages. Fig.10(a) is differential pore volume versus pore size

diameter, and the aperture distribution curve represents the threshold radius. The size of the threshold radius is located in the inflection point region after the horizontal part. Below this radius, the invasion is relatively small. However, once the threshold radius is exceeded, the invasion will start quickly [43]. With the increase of SRA dosage, the aperture distribution moves to the right and the threshold radius increases. In addition, the steep slope in Fig.10(a) indicates the existence of a large number of pores, showing that the pore diameter becomes coarser with the increasing SRA dosage.

Fig.10(b) shows that the cumulative pore volume changes with the pore diameter. It's obvious that the large-pore pore volume of AACGS specimen increases significantly with the increase of SRA dosage. Additionally, there existed little difference in the macroporous pore volume of the specimens when the SRA dosage were 1% and 2%, while both were larger than that of the specimens unmixed SRA. Table 6 shows the pore structure characteristics of AACGS mortar. Obviously, the median diameter and average diameter of mortar specimens increase with the increase of SRA dosage. However, the porosity of overall mortar specimens shows no significant difference. Through carrying out MIP data analysis, it was found that SRA-0 specimen had more pores of 2nm-10nm, while SRA-3 specimen had more pores of 10nm-20nm, which may also be an important reason for the decrease of compressive strength after the addition of SRA [44].



(a) dV/dlogD pore volume (b) Cumulative pore volume
Fig.10 MIP test results of AACGS mortar specimens with different SRA dosage

Table 6 Pore structure characteristics of AACGS mortar

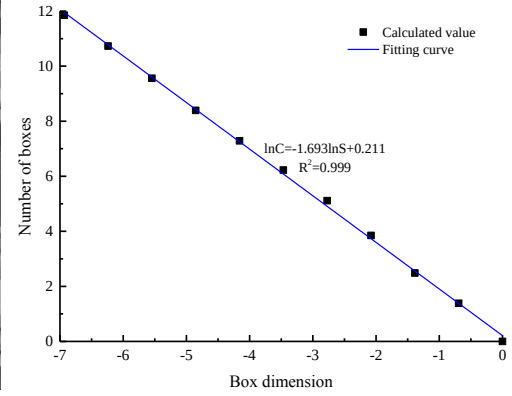
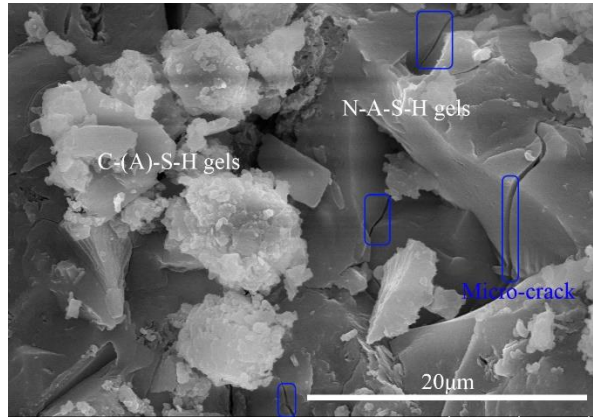
Parameter	SRA-0	SRA-1	SRA-2	SRA-3
Median pore diameter/Volume(nm)	11.6	17.3	19.7	22.7
Average pore diameter/(4V/A)(nm)	10.7	13.6	14.0	16.9
Porosity	17.22%	17.63%	17.48%	17.92%

3.7 SEM analysis

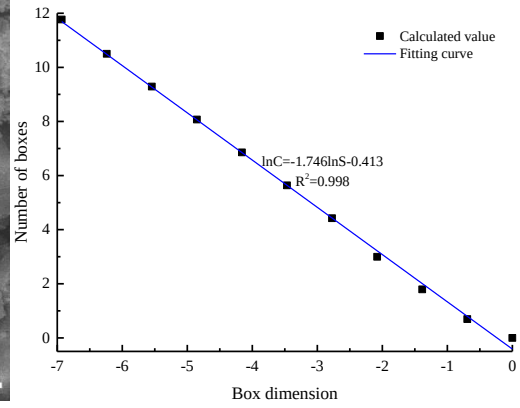
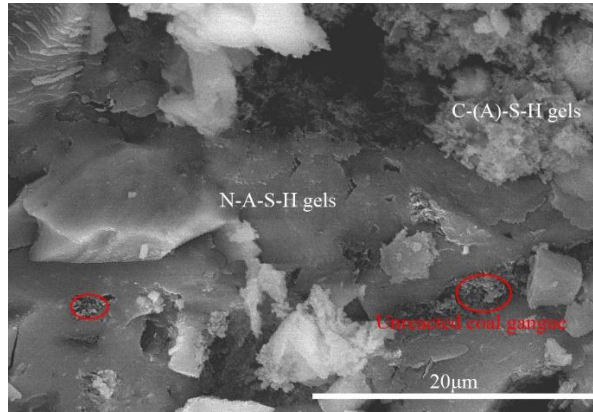
The photos obtained by SEM belong to grayscale images. Since the mixing ratio of AACGS cementitious matrix is the same, the differences of SEM photos of each specimen cannot be quantitatively represented. The fractal dimension can stand for the complexity and roughness of the fractal body, and the larger the fractal dimension, the more complex and rougher the fractal body will be [45]. The rougher and more complex the structure is, the more uneven the pore diameter distribution is, and the larger the fractal dimension is. On the contrary, the simpler the structure is, the more uniform the pore diameter distribution is, and the smaller the fractal dimension is. In the present study, the box dimensions of SEM photos of each specimen are calculated based on Matlab programming. The abscissa is the box dimension, the ordinate is the box number, and the negative value of slope represents the fractal dimension (box dimension). Fig.11 is SEM photos and box dimension calculations of specimens with different SRA dosages. Box dimension values at 0%, 1%, 2% and 3% SRA dosages are 1.693, 1.746, 1.767 and 1.809, separately. That is, the fractal dimension increases with the increase of SRA dosage, indicating that the pore diameter distribution of the specimen structure is more uneven, and the analysis result is the same as that of MIP analysis results. Through analyzing the SEM photos of each specimen in Fig.11, it can be found that there are obvious micro-cracks in SRA-0 specimen. Nevertheless, the micro-cracks width is much smaller than SRA-0 specimen after the addition of SRA. Fractal dimension can not only quantitatively describe the microstructure of different specimens, but also has a good correlation with compressive

strength. Fig.12 shows the relationship between the compressive strength of AACGS concrete and the box dimension. In addition, the compressive strength of AACGS concrete shows a linear attenuation trend when fractal dimension value increases.

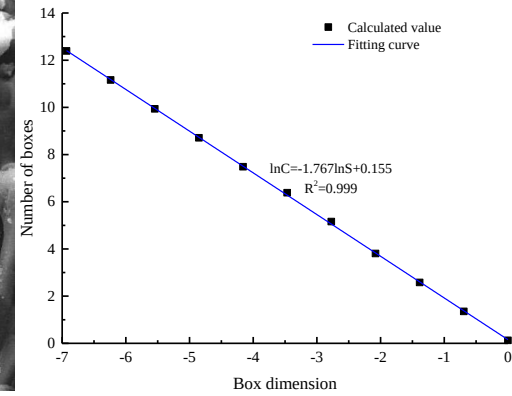
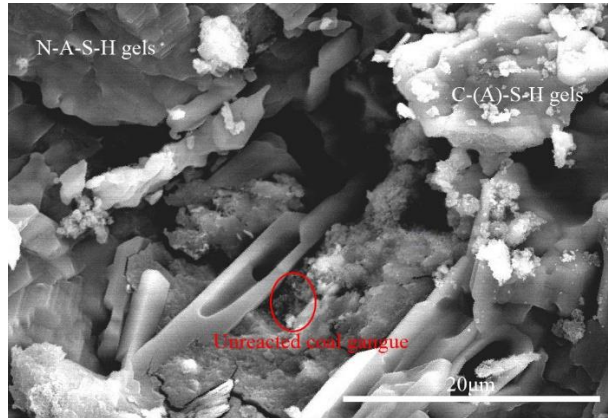
Fig.13 shows SEM photos of AACGS concrete after 0 and 100 freeze-thaw cycles. According to Fig.13(a) and Fig.13(c), micro-cracks in AACGS concrete can be obviously found in specimens unmixed SRA, but no cracks can be found after the addition of SRA. Moreover, compared with the two photos, it is obvious that the ITZ is more compact after the addition of SRA. Fig.13(b) and Fig.13(d) are SEM photos after 100 freeze-thaw cycles. The aggregate morphology in AACGS concrete remains unchanged, and the most fragile place is the bonding interface between the mortar matrix-coarse aggregate after the occurrence of mortar shrinkage. The bonding surface of mortar matrix-coarse aggregate belongs to the weak area, compared with mixing 3% SRA specimens, the width of micro-cracks will be much larger at the bonding surface of mortar matrix-coarse aggregate of specimens unmixed SRA during freeze-thaw. The freeze-thaw damage in the AACGS concrete specimens mixed with SRA was more uniform, and microcracks appeared on both the mortar matrix and the matrix-coarse aggregate bonding surface, and the crack width was smaller than the single crack width in Fig.13(b). It's obvious that the specimens unmixed SRA show macroscopic cracks in ITZ, the width of which can reach up to 9 μ m at most. Compared with SEM photos of specimens with SRA, no obvious wide cracks are found in ITZ. In other words, the addition of SRA can significantly enhance the interfacial bond strength of AACGS mortar and CCGCA, showing stronger resistance to freeze-thaw damage.



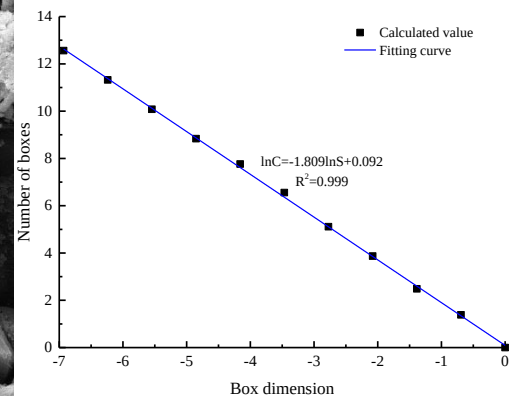
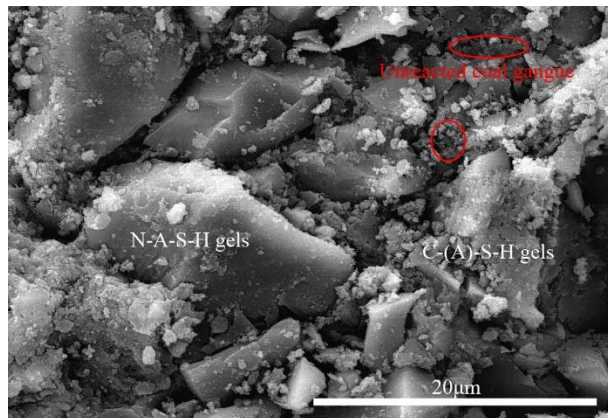
(a) SRA-0



(b) SRA-1



(c) SRA-2



(d) SRA-3

Fig.11 SEM photos and counting box dimension of specimens with different SRA dosages

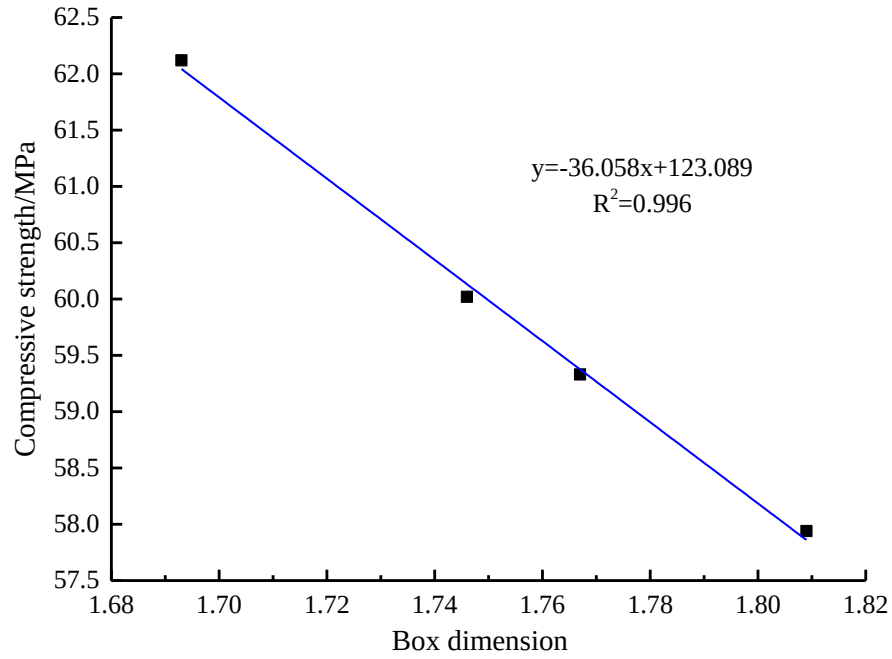
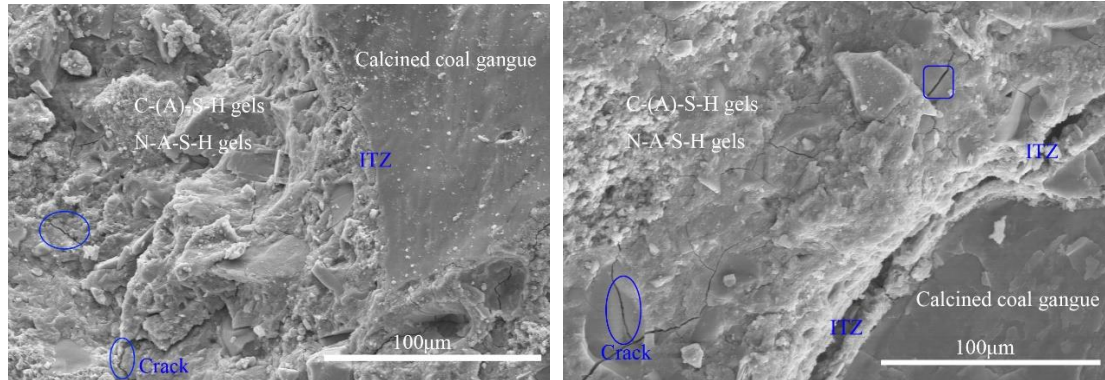
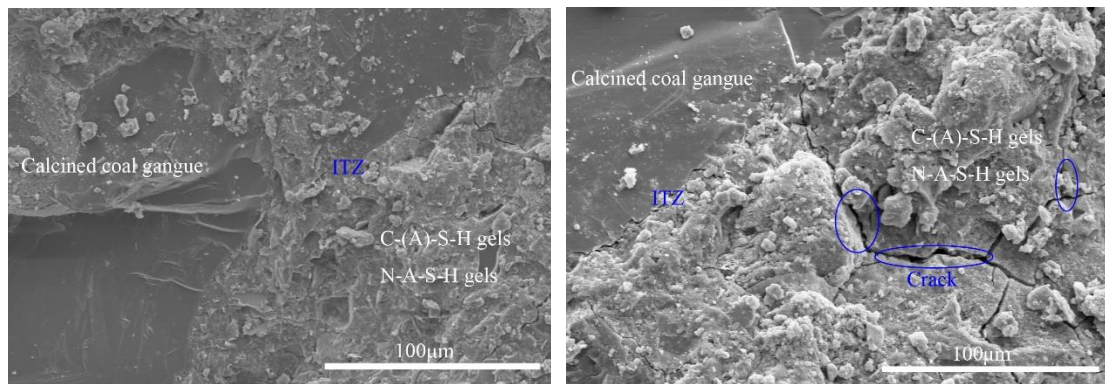


Fig.12 The relationship between the compressive strength of AACGS concrete and the box dimension



(a) Unmix SRA, 0 freeze-thaw cycles

(b) Unmix SRA, 100 freeze-thaw cycles



(c) SRA, 0 freeze-thaw cycles

(d) SRA, 100 freeze-thaw cycles

Fig.13 SEM photos of AACGS concrete after 0 and 100 freeze-thaw cycles

4 Discussion and action mechanism analysis

4.1 Action mechanism analysis of SRA

SRA has a great effect on the polymerization reaction of AACGS mortar. SRA has a strong adsorption effect and forms a strong "organic molecular film" at the water-solid interface, resulting in a reduction in the interfacial energy between the coal gangue-slag particle polymer during the polycondensation process, which hinders the polycondensation process. That is to say, the concentration of alkali ions in the pores of AACGS specimens decreased after the addition of SRA, which reduced the polymerization rate of AACGS pastes in the early stage and led to the decrease of its early strength. With increasing curing age, the alkali environment in the solid still exists, which continues to promote the depolymerization and polycondensation of unreacted slag and coal gangue particles to form stable gelled phase skeleton.

Liu et al. [46] tested the surface tension values of distilled water and distilled water mixed with 1% and 2% SRA, and the test values were 72.3mN/m, 40.6mN/m and 33.8mN/m, respectively, finding that the surface tension values in the mortars pore were lower. Yao et al. [42] reported that higher OH-concentrations promoted the reduction of pore surface tension by SRA. The addition of SRA reduces the polarity of the mixed water, and reduces the affinity of alkali or salt to dissolve and ionize in the mixed water [47]. As a kind of chemical organic solvent, SRA significantly reduces the shrinkage stress caused by capillary water loss during drying, self-drying or curing by reducing the surface tension of pore solution inside concrete, which is also the mechanism provided by all researchers who study SRA to reduce shrinkage [48-50].

Bentz et al. [23] found SRA caused the less evaporation, reduced settlement tension and lower crack inducing stresses at the topmost layer of the mortar. Through carrying out the analysis of the pore structure of the mortar specimens, it was found that the addition of SRA did not significantly

1 481 affect the overall porosity of the specimens. However, it significantly increased the median pore
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3 482 size and average pore size of the mortar specimens. Furthermore, based on the MIP curve analysis,
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6 483 the volume of small pores (2nm-10nm) in specimen SRA-0 was significantly larger. In addition to
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9 484 pore shrinkage and deformation, pore reduction is mainly caused by the collapse and recombination
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12 485 of the gel phase during AACGS polymerization. In fact, the capillary tension stress and pore
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15 486 compression deformation are reduced when SRA dosage increases. Palacios et al. [22] tested the
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17 487 effect of SRA on the hydration heat of alkali-activated slag, finding that the addition of SRA
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20 488 significantly prolonged the hydration induction period, and the large amount of precipitation of the
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23 489 reaction product lasted a long time. SRA acts on the interior of AACGS mortar, resulting in more
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26 490 uniform precipitation and accumulation of the generated products, which can thus reduce the
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29 491 collapse and reorganization of the gelled phase during the curing process, maintain the internal
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32 492 structure relatively stable, and reduce the generation of internal microcracks. In summary, the action
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35 493 mechanism of SRA is a comprehensive effect of reducing pore solution surface tension, reducing
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38 494 the collapse and reorganization of the gelled phase, recombination, and coarsening pore structure.

39 495 **4.2 Analysis of the effect of SRA on the durability of AACGS concrete**

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42 496 The micro-cracks caused by the shrinkage of AACGS are not conducive to the durability of
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45 497 concrete structures. The test results prove that the larger the SRA dosage, the more effective the
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48 498 reduction of shrinkage rate will be. When the concrete used in the test is cured under standard curing
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51 499 conditions, the shrinkage rate will be reduced even more, which can significantly reduce the number
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54 500 and width of micro-cracks generated by the internal shrinkage of AACGS concrete. Different from
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57 501 the compressive strength of concrete, the durability is only associated with the characteristics of
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60 502 mortar and the bonding strength between mortar-coarse aggregate. According to SEM photos, the
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addition of SRA could significantly enhance the ITZ bonding strength of AACGS mortar and CCGCA. The main reason is that the addition of SRA reduces the shrinkage of AACGS mortar. The ITZ between AACGS mortar-CCGCA serves as the weak surface inside the concrete, the shrinkage tension of ITZ is greatly reduced during the curing process. Furthermore, the addition of SRA reduces the early polymerization rate and greatly reduces the collapse and reorganization of the early AACGS mortar, and made the binding of AACGS mortar-CCGCA denser.

The addition of SRA significantly improved the pore structure inside the concrete, and pores with a pore diameter greater than $0.01\mu m$ - $10\mu m$ increase. That is, under the action of the same water expansion pressure, the expansion stress damage caused by small pore diameter is greater than that caused by the large pore diameter. Consequently, the freeze-thaw damage will become more serious. This is the main reason why adding SRA improves the anti-freeze durability of AACGS concrete. Comparing Fig.13(b) and Fig.13(d), there are three main reasons why ITZ is different: Firstly, SRA inhibits the shrinkage of AACGS mortar, which reduces the shrinkage stress of the weak interface, and the bond of mortar matrix-coarse aggregate better during the curing process. Secondly, the viscosity of the pore fluid containing SRA increases and the surface tension decreases, resulting in a reduction in the drive transmission of water in the concrete during the freeze-thaw process. As a result, the micro-cracks of the mortar in the concrete specimens mixed with SRA will generate greater expansion stress in the pores during the freeze-thaw cycle. Thirdly, in the process of freeze-thaw cycles, the freezing temperature of water in small pore size is lower. As a result, the water in large pores freezes first. The ice expansion process squeezes the water inside and disperses it to the nearby small unfrozen pores, resulting in a balance between the pressure inside the large frozen pores and the pressure inside the small unfrozen pores. When the

1 525 temperature reaches the freezing temperature of the small pores, the expansion stress caused by
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3 526 water expansion leads to cracks in the perimeter expansion of the small pores, resulting in damage
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6 527 to the concrete.
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9 528 Pore distribution directly affects the resistance of concrete to chloride ion penetration. The
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11 529 permeability coefficient of ordinary concrete is proportional to the square of the aperture radius and
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14 530 to the first power of the total porosity [51-53]. The beneficial effect of SRA on chloride ion
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17 531 penetration is related to the content of CCGCA. The addition of SRA increases the interface bond
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20 532 between AACGS mortar and CCGCA as well as reduces the number of permeable chloride ions
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23 533 along the bonding interface, and reducing the cracking potential during the curing, which is
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26 534 beneficial to the chloride penetration resistance of concrete. Furthermore, it is found that the
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29 535 chloride ion diffusion coefficients under different SRA dosages are not significantly different from
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31 536 each other through Fig.4(a). That is to say, the non-uniform pore size distribution has little effect on
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34 537 the chloride ion diffusion coefficient of AACGS concrete (without CCGCA). However, the content
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37 538 of CCGCA has a greater impact on the chloride ion diffusion coefficient of AACGS concrete. After
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40 539 mixing CCGCA, a large part of chlorine ion diffusion is carried out at the interface between mortar
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43 540 and CCGCA. In other words, the coordination effect of SRA and CCGCA is mainly to enhance the
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46 541 interface bond of AACGS mortar matrix-CCGCA, which greatly improved the permeability
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49 542 resistance of AACGS concrete to chloride ions.

50 543 Criado et al. [54] found that SO_4^{2-} can serve as an activator to promote the reaction of fly ash
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53 544 to generate N-A-S-H gel and zeolite phase crystals. Compared with calcined coal gangue powder,
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56 545 slag has stronger activity, which can be considered as a nearly complete reaction in alkali-activated
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59 546 reaction. As a result, the interior of AACGS is in low or no calcium state, resulting in very few AFt
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and other expansion substances. When SRA is mixed with concrete, the reaction rate in the early stage of AACGS paste is reduced, resulting in more unreacted powder particles than the unmixed SRA specimens. When soaked in 5% Na₂SO₄ solution, SO₄²⁻ can activate the unreacted coal gangue powder inside, causing an increase in its compressive strength.

5 Conclusions

Based on the above experimental results and action mechanism, the following concluding remarks can be drawn:

(a) The addition of SRA can significantly reduce the drying shrinkage of AACGS mortar, and the drying shrinkage decreases by 47.50% when the SRA dosage is 3%. However, the effect of SRA on the inhibition of drying shrinkage of AACGS mortar is not directly associated with water loss.

(b) The addition of SRA reduced the 28d compressive strength of AACGS concrete, yet did not cause substantial changes in the 90d compressive strength. Furthermore, the addition of SRA significantly improved the anti-freeze durability and sulfate attack resistance (90d testing) of AACGS concrete. With the increase of CCGCA content, the improvement effect of SRA on chloride ion penetration resistance of concrete is more obvious.

(c) The addition of SRA reduced the concentration of alkali ions in the pore solution of AACGS concrete, which reduced the polymerization rate of AACGS paste in the early stage. Besides, SRA did not significantly affect the overall porosity of the specimens, but significantly increased the median pore sizes and average pore sizes of the mortar specimens. SRA acts on the interior of AACGS mortar, resulting in more uniform precipitation and accumulation of the generated products, which can thus reduce the collapse and reorganization of the gel phase during the curing process, keep the internal structure relatively stable, and reduce the generation of internal micro-cracks.

(d) The fractal dimension can be used to quantitatively describe the microstructure of different specimens, and the compressive strength of AACGS concrete shows a negative linear correlation with the increasing fractal dimension.

(e) SRA improves the durability of AACGS concrete through improving the internal pore structure of concrete and increasing the bonding performance of AACGS mortar and CCGCA. In general, the addition of SRA results in the change of internal pore size distribution, reaction rate and interfacial bonding strength of AACGS concrete.

Declaration of Competing Interest

The authors declared that they have no conflicts of interest to this work. We declare that we do not have any commercial or associative interest that represents a conflict of interest in connection with the work submitted.

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References

- [1] A. Fernandez-Jimenez, I. García-Lodeiro, A. Palomo, Durability of alkali-activated fly ash cementitious materials, *J. Mater. Sci.* 42(2017) (9) 3055-3065.
- [2] O. Mikhailova, P. Rovnaník, Influence of polymer admixtures on the rheological properties and heat of hydration of alkali-activated slag pastes, *Solid State Pheno.* 276(2018) 217-222.
- [3] H.L. Ye, Z.J. Chen, L. Huang, Mechanism of sulfate attack on alkali-activated slag: The role of activator composition, *Cem. Concr. Res.* 125(2019) 105868.
- [4] Y. Ma, G. Ye, The shrinkage of alkali activated fly ash, *Cem. Concr. Res.* 68(2015)75-82.
- [5] H.L. Ye, A. Radlińska, Shrinkage mechanisms of alkali-activated slag, *Cem. Concr. Res.* 88(2016) 126-135.
- [6] Z.M. Li, M. Nedeljković, B.Y. Chen, G. Ye, Mitigating the autogenous shrinkage of alkali-activated slag by metakaolin, *Cem. Concr. Res.* 122(2019) 30-41.
- [7] L. Qin, X.J. Gao, Properties of coal gangue-Portland cement mixture with carbonation, *Fuel* 245(2019) 1-12.
- [8] J.Y. Li, J.M. Wang, Comprehensive utilization and environmental risks of coal gangue: A review, *J. Clean. Prod.* 239(2019) 117946-117964.
- [9] Z.C. Dong, J.W. Xia, C. Fan, J.C. Cao, Activity of calcined coal gangue fine aggregate and its

effect on the mechanical behavior of cement mortar, *Constr. Build. Mater.* 100(2015) 63-69.

[10] F. Gao, L.L. Zhao, L.W. Hao, Y. Liu, J.P. Cui, Z.H. Wang, X.Z. Gong, Life cycle assessment of cement clinker production using coal gangue as alternative raw material and fuel, *Mater. Sci. Forum*, 814(2015) 435-440.

[11] X.Y. Yang, Y. Zhang, Z.H. Li, Embankment displacement PLAXIS simulation and microstructural behavior of treated-coal gangue, *Mater.* 10(3) (2020) 218-232.

[12] L.Q. Luo, K.Y. Li, W. Fu, C. Liu, S.Y. Yang, Preparation, characteristics and mechanisms of the composite sintered bricks produced from shale, sewage sludge, coal gangue powder and iron ore tailings, *Constr. Build. Mater.* 232 (2020) 117250.

[13] M. Zhou, Y.W. Dou, Y.Z. Zhang, Y.Q. Zhang, B.Q. Zhang, Effects of the variety and content of coal gangue coarse aggregate on the mechanical properties of concrete, *Constr. Build. Mater.* 220(2019) 386-395.

[14] X. Guan, J.S. Qiu, H.T. Song, Q. Qin, C.H. Zhang, Stress-strain behaviour and acoustic emission characteristic of gangue concrete under axial compression in frost environment, *Constr. Build. Mater.* 220(2019) 476-488.

[15] Y.Z. Zhang, Q.H. Wang, M. Zhou, Y.F. Fang, Z.K. Zhang, Mechanical properties of concrete with coarse spontaneous combustion gangue aggregate (SCGA): Experimental investigation and prediction methodology, *Constr. Build. Mater.* 255(2020) 119337.

[16] H.Q. Ma, H.Y. Chen, H.G. Zhu, Y.Y. Shi, Y.D. Ni, Q.J. Huo, Z.T. Hang, Study on the drying shrinkage of alkali-activated coal gangue-slag mortar and its mechanisms, *Constr. Build. Mater.* 225(2019) 204-213.

[17] L. Kalina, V. Bílek, E. Bartoníčková, J. Krouská, Polypropylene glycols as effective shrinkage-reducing admixtures in alkali-activated materials, *ACI Mater. J.* 115(2018) (2) 251-256.

[18] M.B. Haha, B. Lothenbach, G. Le Saout, F. Winnefeld, Influence of slag chemistry on the hydration of alkali-activated blast-furnace slag-Part I: effect of MgO, *Cem. Concr. Res.* 41(2011) (9) 955-963.

[19] A. Brakat, Y.M. Zhang, Shrinkage mitigation of alkali-activated slag with natural cellulose fibres, *Adv. Cem. Res.* 31(2019) (2) 47-57.

[20] Z.J. Jia, Y.Y. Yang, L.Y. Yang, Y.M. Zhang, Z.M. Sun, Hydration products, internal relative humidity and drying shrinkage of alkali activated slag mortar with expansion agents, *Constr. Build. Mater.* 158(2018) 198-207.

[21] X. Hu, C.J. Shi, Z.H. Zhang, Z.L. Hu, Autogenous and drying shrinkage of alkali-activated slag mortars, *J. Am. Ceram. Soc.* 102(2019) (8) 4963-4975.

[22] M. Palacios, F. Puertas, Effect of shrinkage-reducing admixtures on the properties of alkali-activated slag mortars and pastes, *Cem. Concr. Res.* 37(2007) (5) 691-702.

[23] D.P. Beentz, M.R. Geiker, K.K. Hansen, Shrinkage-reducing admixtures and early-age desiccation in cement pastes and mortars, *Cem. Concr. Res.* 31(2011) (7) 1075-1085.

[24] D. Ballekere Kumarappa, S. Peethamparan, M. Ngami. Autogenous shrinkage of alkali activated slag mortars: basic mechanisms and mitigation methods. *Cem. Concr. Res.* 109(2018) 1-9.

[25] H.Q. Ma, C. Yi, H.G. Zhu, Z.C. Dong, H.Y. Chen, J.X. Wang, D.Y. Li, Compressive strength and durability of coal gangue aggregate concrete, *Mater. Rev.* 32(2018) (7) 2390-2395.

[26] C. Song, Y.C. Choi, S. Choi, Effect of internal curing by superabsorbent polymers-Internal relative humidity and autogenous shrinkage of alkali-activated slag mortars, *Constr. Build. Mater.*

123(2016) 198-206.

[27] V. Bílek, L. Kalina, R. Novotný, J. Tkacz, L. Parížek, Some issues of shrinkage-reducing admixtures application in alkali-activated slag systems, *Mater* 9(2016)(6) 462.

[28] Y.D. Dang, J.S. Qian, Y.Z. Qu, L. Zhang, Z. Wang, D. Qiao, X.W. Jia, Curing cement concrete by using shrinkage reducing admixture and curing compound, *Constr. Build. Mater.* 48 (2013) 992-997.

[29] T. Deboodt, T. Fu, J.H. Ideker, Durability assessment of high-performance concrete with SRAs and FLWAs, *Cem. Concr. Compos.* 57 (2015) 94-101.

[30] L. Coppola, D. Coffetti, E. Crotti, S. Candamano, F. Crea, G. Gazzaniga, T. Pastore, The combined use of admixtures for shrinkage reduction in one-part alkali activated slag-based mortars and pastes. *Constr. Buil. Mater.* 248(2020) 118682.

[31] J.Q. Gong, W. Zeng, W.J. Zhang, Influence of shrinkage-reducing agent and polypropylene fiber on shrinkage of ceramsite concrete, *Constr. Build. Mater.* 159(2018) 155-163.

[32] Y.F. Ling, K.J. Wang, C.Q. Fu, Shrinkage behavior of fly ash based geopolymers pastes with and without shrinkage reducing admixture, *Cem. Concr. Compos.* 98(2019) 74-82.

[33] W.Y. Zhang, Y. Hama, S.H. Na, Drying shrinkage and microstructure characteristics of mortar incorporating ground granulated blast furnace slag and shrinkage reducing admixture, *Constr. Build. Mater.* 93(2015) 267-277.

[34] H.Q. Ma, H.G. Zhu, C. Yi, J.C. Fan, H.Y. Chen, X.N. Xu, T. Wang, Preparation and reaction mechanism characterization of alkali-activated coal gangue-slag materials, *Mater.* 12(2019) 2250.

[35] N. Dz˘unozovic˘, M. Komljenovic˘, V. Nikolic˘, T. Ivanovic˘, External sulfate attack on alkali-activated fly ash-blast furnace slag composite, *Constr. Build. Mater.* 157(2017) 737-747.

[36] Q.F. Liu, G.L. Feng, J. Xia, J. Yang, L.Y. Li, Ionic transport features in concrete composites containing various shaped aggregates: a numerical study, *Compos. Struct.* 183 (2018) 371-380.

[37] M.F. Iqbal, Q.F. Liu, I. Azim, X. Zhu, J. Yang, M.F. Javed, M. Rauf, Prediction of mechanical properties of green concrete incorporating waste foundry sand based on gene expression programming, *J. Hazard. Mater.* 384(2020) 121322.

[38] J. Zhang, X.Z. Weng, L. Jiang, B.H. Yang, J.Z. Liu, Frost resistance of concrete reinforced using surface-strengthening materials in airport pavements, *J. Mater. Civil Eng.* 30(2018) (3) 4018006.

[39] Y. Wang, M.Z. An, Z.R. Yu, S. Han, W.Y. Ji, Durability of reactive powder concrete under chloride-salt freeze-thaw cycling, *Mater. Struct.* 50(2017) (1) 1-9.

[40] I. Ismail, S.A. Bernal, J.L. Provis, S. Hamdan, J.S.J. van Deventer, Microstructural changes in alkali activated fly ash/slag geopolymers with sulfate exposure, *Mater. Struct.* 46(2013) 361-373.

[41] Z. Tang, W.G. Li, G.J. Ke, J.L. Zhou, W.Y. Tam Vivian, Sulfate attack resistance of sustainable concrete incorporating various industrial solid wastes, *J. Clean. Prod.* 218(2019) 810-822.

[42] J. Lee, T. Lee, Effects of high CaO fly ash and sulfate activator as a finer binder for cementless grouting material, *Mater.* 12(2019) (22) 3664.

[43] X.D. Chen, S.X. Wu, Influence of water-to-cement ratio and curing period on pore structure of cement mortar, *Constr. Build. Mater.* 38(2013) 804-812.

[44] Y. Wehbe, A. Ghahremaninezhad, Combined effect of shrinkage reducing admixtures (SRA) and superabsorbent polymers (SAP) on the autogenous shrinkage, hydration and properties of cementitious materials, *Constr. Build. Mater.* 138(2017) 151-162.

[45] X. Yang, F.L. Wang, X.C. Yang, Q. Zhou, Fractal dimension in concrete and implementation

for meso-simulation, *Constr. Build. Mater.* 143(2017) (15) 464-472.

[46] J.H. Liu, F. Nima, C.J. Shi, Shrinkage and strength development of UHSC incorporating a hybrid system of SAP and SRA, *Cem. Concr. Compos.* 97(2019) 175-189.

[47] F. Rajabipour, G. Sant, J. Weiss, Interactions between shrinkage reducing admixtures (SRA) and cement paste's pore solution, *Cem. Concr. Res.* 38(2008) 606-615.

[48] S. de Gemma Rodriguez, G. Arlindo Freitas, Effects of fine LWA and SAP as internal water curing agents, *Int. J. Concr. Struct. Mater.* 8(2014) (3) 229-238.

[49] S. Christof, M. Viktor, G. Michaela, Relation between the molecular structure and the efficiency of superabsorbent polymers (SAP) as concrete admixture to mitigate autogenous shrinkage, *Cem. Concr. Res.* 42(2012) (6) 865-873.

[50] G. Sant, B. Lothenbach, P. Juilland, G.L. Saout, J. Weiss, The origin of early age expansions induced in cementitious materials containing shrinkage reducing admixtures, *Cem. Concr. Res.* 41(2011) (3) 218-229.

[51] Z.J. Shao, J.B. Zhang, J. Liu, W.S. Zhang. Theoretical research on concrete pore fractal characteristic and chloride diffusivity, *Concr.* 275(2012) 1-3.

[52] Q.F. Liu, Z. Hu, X.Y. Lu, J. Yang, I. Azim, W. Sun, Prediction of chloride distribution for offshore concrete based on statistical analysis, *Mater.* 13(1) (2020) 174.

[53] L.X. Mao, Z. Hu, J. Xia, G.L. Feng, I. Azim, J. Yang, Q.F. Liu, Multi-phase modelling of electrochemical rehabilitation for ASR and chloride affected concrete composites, *Compos. Struct.* 207(2019) 176-189.

[54] M. Criadoa, A. Fernández Jiménez, A. Palomob, Effect of sodium sulfate on the alkali activation of fly ash, *Cem. Concr. Compos.* 32(2010) 589-594.