



# Shrinkage mitigation strategies in alkali-activated slag

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## ABSTRACT

In this work, three shrinkage mitigation strategies (i.e. high-temperature curing, sulfate-enrichment, and calcium-enrichment) were evaluated and their effectiveness in mitigating shrinkage of alkali-activated slag (AAS) was studied. The results show that the cause of high-magnitude shrinkage in AAS is attributed to the high visco-elastic/visco-plastic compliance of calcium-alumina-silicate-hydrate (C-A-S-H) in AAS. High-temperature curing can considerably reduce the shrinkage of AAS, likely through strengthening and stabilizing C-A-S-H by improving the coalescence or bonding between adjacent C-A-S-H nanoparticles. However, mitigating shrinkage through early-age expansive reactions is less effective, since the dominant component of drying shrinkage in AAS is due to the long-term visco-elastic/visco-plastic deformation of C-A-S-H.

## 1. Introduction

Ground granulated blast-furnace slag (GGBFS) is a calcium aluminosilicate byproduct of iron ore smelting which has a latent hydraulic reactivity. Alkali-activated slag (AAS) is a promising alternative to ordinary portland cement (OPC), which is typically produced by mixing GGBFS with high alkalinity solution (e.g. sodium hydroxide) to provide strong cementing capabilities [1,2]. The main benefits of AAS relate to the repurposing of industrial byproducts, reducing waste material deposition and requiring little additional energy for clinkering process. However, the industry production of alkalis may limit the potential sustainability of AAS. Nevertheless, recent studies show that AAS concrete provide 25–50% reductions in CO<sub>2</sub> emission and > 40% reductions in embodied energy relative to OPC concrete [2–4]. However, AAS presents several challenges as a construction material, such as high shrinkage, rapid carbonation, potential for alkali-silica reaction, healthy and safety issues associated with handling of an activator, and lack of standard testing [5–10]. All these need to be addressed before broader application of these alternative binders.

### 1.1. Mechanisms of drying shrinkage in AAS

Many studies have shown that AAS undergoes large chemical, autogenous, and drying shrinkage, which can result in cracking and loss of durability and serviceability in restrained concrete elements. The drying shrinkage of AAS was measured to be up to six times higher than shrinkage of OPC [11–13]. The cause of the high-magnitude shrinkage in AAS, however, is not completely understood. At high relative

humidity (RH), the generation of capillary pressure due to meniscus formation in pores is commonly regarded as the main driving force for shrinkage in cementitious materials [14–16]. The correlation among RH above the meniscus, radius of meniscus  $r_c$  in a cylindrical pore, and the capillary pressure can be expressed by Kelvin-Laplace equation [14,17]:

$$p_c = \rho_l \frac{RT}{M} \ln \frac{RH}{a_w} = - \frac{2\gamma \cdot \cos \theta}{r_c - t} \quad (1)$$

where  $p_c$  is the capillary pressure [Pa],  $\rho_l$  is density of pore liquid [g/m<sup>3</sup>],  $M$  is molar mass of pore liquid [g/mol],  $R$  is the universal gas constant [(8.314 J/(mol K))],  $T$  is temperature [K],  $a_w$  is the water activity of pore solution [unitless],  $r_c$  is the capillary radius at the position of meniscus (also named as the Kelvin radius) [m],  $\gamma$  is the surface tension between pore water and vapor [N/m],  $\theta$  is the contact angle denoting the hydrophilicity of the pore wall [°], and  $t$  is the thickness of an adsorbed layer [m], which is a function of RH [18].

The Kelvin radius is also the radius of the largest water-filled cylindrical pore at equilibrium condition [19]. In other words, all surface pores whose radii are smaller than the Kelvin radius are saturated, whereas larger surface pores are dried and only contain a layer of adsorbed water with a thickness of  $t$ . As such, the shrinkage of a partially saturated porous elastic medium due to capillary pressure in the water-filled pores can be approximated as [20,21]:

$$\epsilon_{cap} = \frac{S_w p_c}{3} \left( \frac{1}{K} - \frac{1}{K_s} \right) \quad (2)$$

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**Table 1**  
Oxide compositions of the GGBFS (by mass %).

	CaO	SiO <sub>2</sub>	Al <sub>2</sub> O <sub>3</sub>	MgO	SO <sub>3</sub> <sup>a</sup>	S <sup>2-</sup>	Fe <sub>2</sub> O <sub>3</sub>	Na <sub>2</sub> O	K <sub>2</sub> O	P <sub>2</sub> O <sub>5</sub>	MnO	LOI
Slag	43.83	30.04	12.74	4.79	3.11	0.85	1.16	0.24	0.40	0.08	0.22	2.56

<sup>a</sup> The high sulfate content is attributed to the presence of gypsum in raw GGBFS.

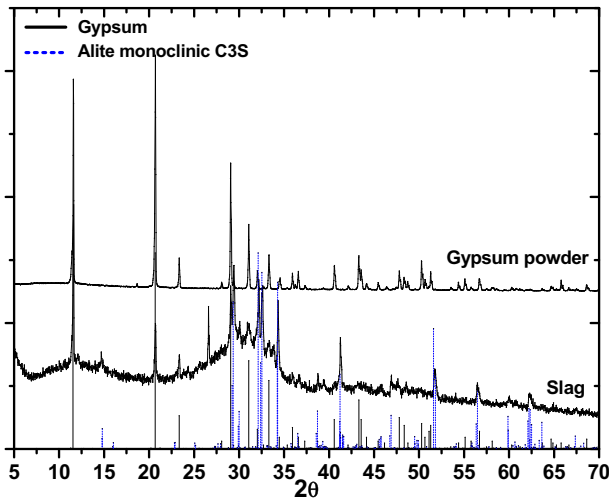


Fig. 1. XRD patterns of GGBFS and gypsum powders.

In which  $\epsilon_{cap}$  is the linear elastic shrinkage,  $K$  is the bulk modulus of the whole porous body [Pa],  $K_s$  is the bulk modulus of the solid material [Pa].  $S_w$  is the degree of saturation, and  $S_w p_c$  is the effective capillary stress. It should be noted that Eq. (2) is only strictly applicable to fully saturated linear elastic porous materials, and approximate for partial saturation cases [22].

Based on Eq. (2), the large drying shrinkage of AAS, in comparison to OPC, can be attributed to (i) a higher degree of saturation at a given RH; (ii) degraded bulk modulus of AAS; (iii) high visco-elastic/visco-plastic compliance of AAS or its solid skeleton. A high degree of saturation in a porous material can be related to a refined pore structure and low water activity of pore solution. In regards to explaining the cause of high shrinkage in AAS, some researchers argued that a high effective capillary stress builds in AAS during drying due to its fine pore size distribution [12]. However, recent studies suggested that the large shrinkage of AAS is attributed to the high visco-elastic/visco-plastic compliance (low creep modulus) of the solids formed in AAS due to alkali enrichment [23,24]. It is argued that the alkali-enriched calcium-alumina-silicate-hydrate (C-A-S-H) in AAS is thermodynamically unstable and tends to reorganize and redistribute upon drying-induced internal stresses [23]. In fact, drying shrinkage of hardened OPC paste mixed with high alkalinity solution exhibits comparable shrinkage magnitude and kinetics, as that in AAS [25].

### 1.2. Mitigation of drying shrinkage in AAS

According to the aforementioned shrinkage mechanisms of AAS, several pertinent mitigation strategies can be developed in AAS. Based on the mitigation mechanisms, the strategies can be roughly categorized into four types: (1) Reducing the impact of shrinkage driving forces; (2) Early-age expansive reactions; (3) Elevated temperature treatment; (4) Internal restraint.

#### 1.2.1. Reducing the impact of shrinkage driving forces

Considering that capillary pressure is the dominant shrinkage driving force for AAS at high RH [12,16], mitigation strategies can be developed through either reducing the surface tension of pore liquid

**Table 2**  
Mix proportion for AAS pastes.

Parameters	Mixture ID <sup>a</sup>				
	Control/HT	GY5	GY10	CaO5	CaO10
NaOH dry pellets (g)	47.5	47.5	47.5	47.5	47.5
Water (g)	249.9	249.9	249.9	249.9	249.9
Slag (g)	603.9	573.7	543.51	573.7	543.51
Gypsum (g)	0	22.57	45.14	0	0
CaO (g)	0	0	0	34.79	69.58
(Gypsum or CaO)/solid <sup>b</sup> (by volume)	5.0%	5.0%	10.0%	5.0%	10.0%
(Gypsum or CaO)/solid (by mass)	0.0%	3.8%	7.7%	5.7%	11.3%
Liquid (activator)/solid (vol./vol.)	1.3	1.3	1.3	1.3	1.3
Liquid (activator)/solid (wt./wt.)	0.49	0.50	0.51	0.49	0.49

<sup>a</sup> In the nomenclature, HT represents AAS with high-temperature curing, GY represents AAS blended with gypsum, CaO represents AAS blended with CaO.

<sup>b</sup> The solid means the sum of raw slag and gypsum or CaO powder.

**Table 3**  
Summary of the experimental program for AAS characterization<sup>a</sup>.

Mixture ID <sup>b</sup>	During length and weight measurements		In sealed vials/containers	
	Before drying (24 h)	Drying at 70% RH (nitrogen purged)	23 ± 0.5 °C	80 ± 0.5 °C
Control	XRD, NST, ICP-AES	XRD (28 d), NST (7 d, 28 d, 70 d)	MIP (28 d), SEM (28 d)	SEM (28 d), XRD (1 h, 1 d, 7 d, 28 d)
GY5	XRD	– <sup>c</sup>	–	XRD (1 h, 1 d, 7 d, 28 d)
GY10	XRD, ICP-AES	XRD (28 d)	MIP (28 d)	
CaO5	XRD	–	–	
CaO10	XRD, ICP-AES	XRD (28 d)	MIP (28 d)	
HT	NST, ICP-AES	NST (7 d, 28 d, 70 d)	MIP (28 d)	–

<sup>a</sup> The abbreviation of experimental techniques can be found in the titles of Sections 2.4–2.8.

<sup>b</sup> The difference between HT mixture and Control mixture at 80 ± 0.5 °C is that HT mixture was cured at 80 ± 0.5 °C merely for the first 16 h, while the latter one was continuously cured at high temperature.

<sup>c</sup> Not tested.

[26] or implementing internal curing [9]. The introduction of shrinkage-reducing admixture (SRA) reduces the surface tension of pore solution by half, and hence reduces the Kelvin radius at a given RH, according to Eq. (1) [27–29]. Therefore, in comparison to the plain system, the system containing SRA exhibits a slightly lower degree of saturation and higher mass loss [27,30,31]. As such, the effective capillary stress, decreases since the capillary pressure is primarily controlled by the RH at equilibrium condition. Although the appropriate incorporation of SRA can reduce the drying shrinkage of AAS up to approximately half of that measured for plain system, the shrinkage magnitude is still considerably high in comparison to OPC [26].

Internal curing is a process, by which saturated porous inclusion (e.g. superabsorbent polymer, lightweight aggregate) is added to the

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