Effects of Solid Activator and Fly Ash on Rheology and Thixotropy of One-Part Alkali-Activated Pastes

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Abstract

Compared with conventional two-part alkali-activated materials (AAMs), one-part AAMs have several advantages, especially the simple production using only water similar to ordinary Portland cement. However, one-part AAMs are much less researched, especially in terms of rheology. Understanding the rheological behavior of such binders is a crucial step for their production, placement, and application in the construction industry. This paper studies the rheological properties of one-part AAMs through evaluating the dynamic yield stress, static yield stress, and thixotropic index. The effects of solid activator content, besides the effect of two types of fly ash (containing different amounts of carbonaceous impurities) with varying contents, are explored. The possible mechanisms are also discussed.

1. Introduction

The trend created by alkali-activated materials (AAMs) in the research sector is prospering, aiming to provide a sustainable “toolkit” of binders via utilizing several precursors (Khalifa et al. 2020; Coffetti et al. 2022) to replace ordinary Portland cement (OPC) partially. The use of blends of fly ash and slag in specific ratios for the production of AAMs has proved to be an innovative approach to achieve similar properties to OPC and magnesium potassium phosphate cement (MKPC) (Matalkah and Soroushian 2018; Shang et al. 2018). Most of the studies on AAMs paid attention to the “two-part” terminology using liquid-based activators, while the "one-part" AAMs using solid-based activators have been less researched (Luukkanen et al. 2018).

Generally, the use of liquid-based activators to produce two-part AAMs can yield a relatively better mechanical performance compared with the solid activators in one-part AAMs (Alrefaei et al. 2021). Yet, the handling, transportation, and operation of liquid-based alkali activators are challenging in in-situ construction applications, which jeopardize the safety of worksites (Luukkanen et al. 2018; Provis 2018; Alrefaei et al. 2019). On the other hand, the technology of one-part AAMs has excellent potential due to its simple operation, scalable cast-in-situ applications, and distributivity as bagged material (Luukkanen et al. 2018; Provis 2018; Matalkah and Soroushian 2019). Hence, several research studies have implemented different approaches to improve the mechanical properties of one-part AAMs (Dong et al. 2020; Almakhadmeh and Soliman 2021; Alzaza et al. 2021; Gökçe et al. 2021). However, the rheological behavior of such binders still needs further research prior to their wide-range in-situ implementations, either as building or repairing materials.

Several studies focused on the rheological behavior of the two-part AAMs, including pastes (made from fly ash (Rashad et al. 2014; Palacios et al. 2011; Favier et al. 2014), slag (Rashad 2013; Puertas et al. 2014; Palacios et al. 2021), metakaolin (Poulesquen et al. 2011; Favier et al. 2014), or their blends (G et al. 2019; Guo et al. 2020), mortars (Alonso et al. 2017), and concrete (Puertas et al. 2018). In contrast, limited studies focused on one-part AAMs (Luukkanen et al. 2019; Li et al. 2020; Lu et al. 2021; Alrefaei and Dai 2022). According to the authors’ previous study (Alrefaei et al. 2021), the two-part AAMs might exhibit a comparatively modest thixotropic behavior relative to the one-part AAMs. Another study confirmed the weak thixotropic behavior of the two-part AAMs (using metakaolin and sodium silicate solution) compared to OPC due to the absence of colloidal interaction (Favier et al. 2014). Nevertheless, limited studies implemented the hysteresis test and stress decay method to observe the thixotropic behavior of two-part AAMs (Panda et al. 2018; Kondepudi and Subramaniam 2019) and one-part AAMs (Guo et al. 2020), while other studies (Panda et al. 2019; Bong et al. 2021; Muthukrishnan et al. 2021) reported the viscosity recovery behavior besides the static yield stress of both one-part and two-part AAMs for 3D printing purposes. Recent studies (Dai et al. 2020a, 2020b) reported the

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effect of activator properties and water content on the structural build-up behavior of alkali-activated fly ash/slag pastes. Some research sheds light on the thixotropic behavior of one-part fly ash/slag AAMs based on flow table test visual observations (Nematollahi et al. 2017; Alrefaei et al. 2019, 2020), yet no systematic study has been done.

Thixotropy can be defined as the change in apparent viscosity of the material, in which the viscosity drops upon shearing and rises upon shear removal over time (Qian et al. 2018). In general, the rheological properties of thixotropic materials are theoretically reversible, isothermal, and time-dependent; consequently, they exhibit dynamic and static yield stresses (Qian and Kawashima 2018). According to previous research, thixotropy can be evaluated using several techniques such as hysteresis tests (Assaad et al. 2003; Ferron et al. 2007), shear rate decay method (Kawashima et al. 2013), and shear stress decay under constant shear rate (Roussel 2006; Qian and Kawashima 2016a). The typical shear stress behavior of cement under a constant intermediate shear rate starts from a rapid increase to a maximum value (maximum shear stress needed to initiate flow, \( \tau_x \)) followed by gradual decay until reaching equilibrium value (steady-state flow, \( \tau_r \)). Such a response is related to thixotropy (Qian and Kawashima 2018). Several models were proposed to describe the exponential decay of shear stress over time and evaluate thixotropy (Roussel 2006). The thixotropic index was proposed in previous research to assess thixotropy, calculated as a ratio between \( \tau_x \) and \( \tau_r \) (Qian et al. 2018). The term "characteristic time" is typically used to represent the destructuration rate of the fresh cement particles network (Qian et al. 2018). A shorter characteristic time reflects a faster destructuration rate. Meanwhile, \( \lambda_{diss} \), which characterizes the increasing rate of static yield stress over a relatively long period of time (thousands of seconds), has also been popularly used to quantify the thixotropy of cementitious materials (Roussel 2006).

Thixotropy is of great importance for several engineering applications like self-compactend concrete (SCC), formwork pressure, and additive manufacturing (3D printing) (Kim et al. 2010; Qian and Kawashima 2016b; Lu et al. 2019). Accordingly, this study aims to clarify the rheology and thixotropy of one-part alkali-activated fly ash/slag pastes. The effects of solid activator content, fly ash content, and fly ash composition on the dynamic yield stress, static yield stress, and stress decay process are delivered in this work.

2. Materials

2.1 Raw precursors and solid activator

Two different types of fly ash (denoted as FA1 and FA2) were used in this study. FA1 is a local commercial product in Hong Kong produced by Green Island Cement Co. Ltd., while FA2 is collected from China Light & Power Group. It is worth highlighting that the FA1 is produced in a temperature range of 800-900°C using circulating fluidized bed combustion (CFBC) technology, while the FA2 is generated by pulverized coal combustion (PCC) technology that implements a temperature range of 1300–1700°C (Wang et al. 2020). Ground granulated blast-furnace slag (GGBS, shortcutted as "S") powder was also used in this study, as delivered from mainland China.

The solid activator used in this study was anhydrous sodium metasilicate (\( \text{Na}_2\text{SiO}_3\)-Anhydrous) powder, with a chemical composition of 50.46% \( \text{Na}_2\text{O} \) and 47.24% \( \text{SiO}_2 \) by weight (the modulus ratio \( M_s = \text{SiO}_2/\text{Na}_2\text{O} = 0.94 \)), a bulk density of 1.22 g/cm\(^3\), and a solubility in water of 210 g/l at 20°C as provided by the supplier (International Laboratory, USA). The \( D_{50} \) was 679.5 μm, as tested by a laser particle size analyzer. Such an activator is commonly used in the synthesis of one-part AAMs that results in thixotropic pastes, according to previous research (Nematollahi et al. 2015; Alrefaei and Dai 2018).

2.2 Material characteristics

- **Morphologies:** The morphologies of the raw precursors were observed using a scanning electron microscope (SEM) (VEGA3 Tescan), as shown in Fig. 1. The FA1 (Fig 1a) included a high content of irregular shapes of unburnt carbon (briefed as "impurities" in the discussion), a limited amount of shell-like cenospheres, rough clusters of microspheres, and the aluminosilicate sphere particles. The availability of such impurities and bio-contaminations is related to the relatively lower temperature (800-900°C) in the combustion zone (Yang et al. 2018) and the composition of raw coal during production. Such impurities have a porous nature with a relatively high specific surface area (SSA) (20 to 80 m\(^2\)/g for unburnt carbon), which demands a high content of water during mixing (Zhang et al. 2016; Yang et al. 2018). The FA2 (Fig 1b) was of high sphericity with the absence of irregular shapes due to the high production temperature that eliminates the unburnt carbon residues. Finally, the GGBS was mostly of anomalous shapes, as commonly reported in previous research.

- **Chemical and mineralogical compositions:** Table 1 summarizes the chemical composition of the raw precursors. The raw materials were further analyzed through the semiquantitative X-ray diffraction (QXRD) method (each sample was mixed with 20% \( \text{Al}_2\text{O}_3 \) by weight) via the Rietveld refinement method. The non-crystalline (amorphous) phases in the FA1, FA2, and GGBS were 75.66\%, 84.65\%, and almost 100\%, respectively. The FA1 contained 17.26% of quartz, 3.9% of hematite, 1.28% of mullite, and 1.9% of calcite, while the FA2 comprised 9.64% of quartz and 5.71% of mullite. The glassy phases were highlighted in a previous study as a factor that might affect the reactivity and strength development of FA-AAMs (Fernandezjimenez...
et al. 2006). The loss on ignition (LOI) results of both FA1 and FA2 were 5.55% and 1.12%, respectively.

- **Physical characteristics:** The physical properties of raw materials, including the bulk density, particle size distribution (PSD), and SSA, are presented in Table 1. The bulk density of the raw precursors was measured in accordance with ASTM C188, where kerosene was the liquid medium. As shown in Table 1, the FA1 had a lower density relative to that of FA2, which is in agreement with LOI results. The higher impurity content in FA1 is the main reason for such lower density and more significant LOI. Figure 2 shows the PSD of the raw precursors, wherein the GGBS and FA2 showed a relatively similar distribution, while the FA1 included relatively larger sizes (D<sub>50</sub> results are reported in Table 1). On the other hand, the accessible SSA (measured by the Brunauer-Emmett-Teller (BET) method) of FA1 was the highest among all raw materials, although it had relatively larger particles. Further, the accessible SSA of FA2 was slightly lower compared with that of GGBS. Previous research (Zhang et al. 2016; Yang et al. 2018) confirmed that the SSA is of great importance to understanding the rheological behavior. The mass of carbonaceous particles available in the FA1 and FA2 was 1.1% and 0.42%, respectively. Besides their rough morphology, such carbonaceous particles profoundly affect the SSA and water demand of the fly ash (Zhang et al. 2016).

3. Methodology

3.1 Mix proportions

The mix proportions used in this study are presented in Table 2. This study is divided into two parts as follows: 1) the effect of solid activator content, and 2) the effect of fly ash type and content. In the first part of this study, the binder is fixed to be a blend of 50% FA1 and 50% GGBS, while the only variable is the solid activator content ranging between 4% to 10% by the total mass of binder. Such a range of solid activator content complies with the recommendation of previous research (Nematollahi et al. 2015; Criado et al. 2018). It is good to mention that the water-to-binder ratio (w/b) is also fixed at 0.45. However, due to the change of solid activator content in each mix, the liquid-to-solid ratio (l/s) cannot be fixed, where “liquid” is the water and “solid”
is the mass of slag, fly ash, and anhydrous sodium metasilicate. Such liquid-to-solid ratio (l/s) is dependent on the solid activator content (l/s is 0.409, 0.417, 0.425, and 0.433 for the solid activator contents of 10%, 8%, 6%, and 4%, respectively). Consequently, the authors decided to fix the w/b instead of l/s as it was assumed that all the solid activator particles would be dissolved during the relatively long mixing procedure.

In order to study the effect of fly ash on the rheological properties of one-part AAM, the solid activator and w/b are fixed at 10% and 0.45, respectively, and various replacement ratios of fly ash over GGBS are studied. Six different precursor combinations are implemented as follows: 1) 55% FA1 + 45% GGBS, 2) 50% FA1 + 50% GGBS, 3) 20% FA1 + 80% GGBS, 4) 100% GGBS, 5) 50% FA2 + 50% GGBS, and finally 6)100% FA2.

The codified mix ID contains three alphanumeric character parts. The first part refers to the fly ash content and type: "FA1 and FA2 to distinguish the fly ash type", while the second part shows the GGBS content used in the mix design. Finally, the third part denotes the activator content used in the mix.

### 3.2 Zeta potential measurements

Zetasizer Nano ZS (Malvern) was used to measure the
zeta potential of 50/50 FA1 and GGBS blended suspensions to demonstrate the effect of activator content on the rheology of one-part AAMs. The suspensions were dispersed in MilliQ water (0.5 wt.% suspension). Five suspensions were prepared, at which four included different activator contents varying from 4% to 10%, and one was made with no activator as a benchmark reference. All the samples were placed in a sonicator for 10 minutes before the measurement. Three measurements were taken for each suspension.

3.3 Mixing procedure and sample preparation
The mixing procedure was fixed to ensure that all the samples were prepared in the same conditions. All the samples were prepared using tap water and mixed with a 450 watts egg mixer. For each mix, all the solid ingredients were dry mixed by hand for one minute in a plastic cup. After adding the full amount of mixing water to the powder, all the ingredients were mixed at high speed of 720 rpm for 2 minutes. Then, the mixture was left to rest for 6 minutes. After resting, the paste was mixed for another 2 minutes at high speed of 720 rpm. Immediately after 10 minutes of mixing, 35 grams of the paste was weighed in the measuring cup, and then the cup was fixed on the rheometer within 30 seconds before the beginning of the testing protocols. All the tests were conducted at room temperature of 24 ± 2°C.

3.4 Rheometer and construction cell
The rheometer used in this study is an AMETEK BROOKFIELD RST-CC Rheometer. The rotor is a 4-bladed vane with a diameter of 20 mm and a height of 40 mm, while the cup is a custom-made grooved shape with 24 profiles distributed evenly along the wall. Each groove is 2 mm in width and 1 mm in depth. The inner and outer diameters of the cell are 22 mm (from the groove face to face) and 24 mm, respectively, while its height is 65.5 mm. Such a design will ensure the prevention of the wall-slip phenomenon (Qian and Kawashima 2016a). The gap between the bottom of the rotor and the bottom of the cup is 3 mm. The rheometer, cup, and rotor are displayed in Fig. 3.

3.5 Rheology protocols
The one-part AAMs are thixotropic binders; consequently, their rheological behavior is highly susceptible to the flow history. Roussel et al. (2019) assured the importance of implementing a rigorous procedure to ensure the reproducibility of results. From here comes the importance of preshearing to confirm that the same destructed state is achieved for all samples before testing (Feys et al. 2017). Accordingly, all the samples in this study were presheared at an angular velocity of 100 rad/s for 5 minutes, followed by 2 minutes resting before launching the testing protocols. The data acquisition rate was one data point per second. Three new samples were tested for each mix and protocol. Three rheological protocols are applied in this study, summarized as follows:

• **Equilibrium flow curve and dynamic yield stress:** an angular velocity step-down protocol, from 100 rad/s to 90, 80, up till 20 rad/s for 20 seconds each, is used. The average shear stress value was calculated at each angular velocity step and reported with an error bar in the figures. The obtained equilibrium flow curves are fitted into the modified Bingham model (Yahia and Khayat 2001):

\[
\tau = \tau_y + \eta_p \dot{\gamma} + c \dot{\gamma}^2
\]  

where \(\tau_y\) is the dynamic yield stress, \(\eta_p\) is the plastic viscosity, and \(c\) is a constant. Each sample was fitted separately, then the average values of the dynamic yield stress were reported in Tables 3 and 4.

• **Static yield stress:** an intermediate constant angular velocity of 0.011 rad/s (equivalent to a shear rate of 0.125 1/s) is applied for 5 minutes, and the torque development is recorded. The typical response of a material under an intermediate constant angular velocity is that the torque rises to a peak value and finally drops to an equilibrium value over time (Qian and Kawashima 2018). Such a peak value is correspondent to the static yield stress (Mahaut et al. 2008).

• **Thixotropic index and characteristic time:** a constant angular velocity of 100 rad/s (equivalent to a shear rate of 1150 1/s) is applied for 60 seconds to evaluate the torque decay process. Under a constant shear rate,
there is a stress decay corresponding to the destructuration process. Empirical and theoretical studies (Papo 1988; Roussel 2006) have found that a simple exponential model could fit the yield stress decay of pastes very well:

\[ \tau = \tau_s + (\tau_i - \tau_s) e^{-\alpha \gamma t} \]

where \( \tau_s \) and \( \tau_i \) are the initial and equilibrium shear stresses of the decay curve, respectively, while \( \alpha \) is a constant directly related to shear rate (\( \gamma \)) and material properties. The thixotropic index is calculated as \( I_{\text{thix}} = \frac{\tau_s}{\tau_i} \), which has been defined to quantify the thixotropy (Qian and De Schutter 2018; Qian et al. 2018). The characteristic time of the deflocculation is calculated as \( \frac{1}{\alpha \gamma} \), which indicates how fast the destruction process is.

### 4. Results

#### 4.1 Effect of solid activator content

**4.1.1 Equilibrium flow curve and dynamic yield stress**

The equilibrium flow curves of the one-part AAMs incorporating various activator contents are plotted in Fig. 4. The dynamic yield stress results obtained from the model fitting of the equilibrium flow curves and the plastic viscosity results are reported in Table 3, at which the \( R^2 \) values for all series were 0.999. It is observed that the increase of the solid activator content in the one-part AAMs slightly increased the dynamic yield stress of the one-part AAMs. Moreover, as shown in Fig. 4, the slope of flow curves slightly increased with the increase of solid activator content, which is directly related to the plastic viscosity of the paste. Previous research (Puertas et al. 2014; Palacios et al. 2019) confirmed that the activator concentration contributes to higher yield stress and apparent viscosity of the two-part alkali-activated pastes. To sum up, the dynamic yield stress was increased by 5%, 37%, and 73% for 6%, 8%, and 10% of activator addition compared with that of 4% activator usage.

**4.1.2 Static yield stress**

The shear stress development results of the one-part AAMs with various activator contents under 0.011 rad/s are displayed in Fig. 5, wherein the error bars were excluded to maintain clarity. The higher the activator content, the higher both the peak and equilibrium shear stress values would be. The peak values of all the samples are reported in Table 3, which correspond to the static yield stress. The higher the peak, the higher the static yield stress will be. The activator content markedly influenced the static yield stress of the one-part AAMs, at which the increase of activator content exponentially boosted the static yield stress. To sum up, the static yield stress was increased by 48%, 214%, and...
780% when using 6%, 8%, and 10% solid activator contents, respectively, relative to that of 4% activator content. It is worth noting that an increase in shear stress was observed in some series beyond 150 seconds, which might be attributed to the precipitation of new reaction products (Puertas et al. 2014; Palacios et al. 2021).

4.1.3 Thixotropic index and characteristic time
The torque decay performances of the one-part AAMs with various activator contents are presented in Fig. 6. The solid activator content directly affected both peak and equilibrium values of the stress decay response, at which increasing the activator content had increased both values. The gap between peak and equilibrium values (τ_p - τ_e) increased with the increase of the utilized activator content.

The thixotropic index (Ithix) and characteristic time of destructuration results are plotted in Figs. 7a and 7b, respectively. As shown in Fig. 7a, the thixotropic index (Ithix) was relatively similar in the range between 4% to 6% activator content, then significantly increased with activator content. Such an increase was more noticeable from 8% to 10% activator content relative to that from 6% to 8%.

As shown in Fig. 7b, the characteristic time dropped with the increase of solid activator. This might indicate the higher interconnectivity and bonding between the precursor particles of the alkali-activated paste when higher activator content is used. Such interconnectivity between the precursor grains might be related to the inter-particle adhesion forces, as will be discussed in section 5.1.

4.2 Effect of fly ash type and content
4.2.1 Equilibrium flow curve and dynamic yield stress
Figure 8 presents the equilibrium flow curves of the one-part pastes incorporating different types and contents of fly ash, and the calculated dynamic yield stresses and plastic viscosity results are reported in Table 4, at which the R² values for all series were 0.999 except FA2 series with R² values of 0.939. In reference to the fly ash-free AAM paste (0FA/100S-10%), the addition of
FA1 increased the plastic viscosity of the one-part AAMs, while the incorporation of FA2 resulted in reducing the plastic viscosity of their corresponding pastes. The higher the FA1 content in the one-part AAMs, the higher the dynamic yield stress of fresh pastes. On the other hand, the more the included FA2 content in the one-part pastes, the lower the dynamic yield stress. It is worthy of highlighting that the dynamic yield stresses of the FA2 pastes were very low to the extent that their values were close to zero. In summary, the dynamic yield stress was increased by 21%, 50%, and 139% when including 20%, 50%, and 55% of FA1 in the AAMs, yet it dropped by 91% and 92% for 50% and 100% replacement of FA2, respectively, relative to fly ash-free pastes.

4.2.2 Static yield stress

The effect of fly ash type and content on the shear stress development of the one-part alkali-activated pastes under 0.011 rad/s is shown in Fig. 9, and the peak values of the response curves are reported in Table 4, referring to the static yield stress. The addition of FA1 was found to increase the static yield stress of the one-part AAMs significantly. In contrast, the static yield stress of the AAMs significantly dropped when including FA2 till almost reaching zero for the 100% FA2 AAM paste. In summary, the static yield stress was increased by 9%, 73%, and 356% when 20%, 50%, and 55% of FA1 was incorporated in AAMs, yet it dropped by 70% and 95% for 50% and 100% replacement of FA2, respectively, relative to fly ash-free AAM pastes.

4.2.3 Thixotropic index and characteristic time

Figure 10 shows the stress decay curves of the one-part AAMs with different fly ash types and contents under 100 rad/s. The addition of FA1, to a certain extent, contributed to higher $\tau_i$ and $\tau_e$ values. Contrarily, the higher the FA2 content, the lower both peak and equilibrium values would be. It is good to mention that the FA2 pastes were not thixotropic in this study, which explains the absence of clear initial peaks in their stress decay responses in Fig. 10. The torque decay responses of the 50%-FA1 and 55%-FA1 were relatively similar; however, the initial peak value of the 55%-FA1 AAM was 23% higher compared with that of the 50%-FA1 AAM. This might highlight the effect of the FA1 content on the thixotropy of one-part AAMs.

The effect of fly ash type and content on the thixotropic index ($I_{thix}$) and characteristic time of deconstruction of the one-part AAMs at 100 rad/s are plotted in Figs. 11(a) and 11(b), respectively. In reference to the FA-free AAM pastes (OFA/100S-10%), the thixotropic index dropped after the addition of a relatively low amount (20%) of FA1, which might be related to the ball-bearing and lubricating effect of the FA particles that weakened the bonding between particles/agglomerates. Increasing the FA1 content up to 50% caused an increase in the thixotropic index, yet the average value was still comparatively lower relative to that of the FA-free paste. However, for FA1 content of 55%, the thixotropic index surpassed that of FA-free
paste. Conversely, the incorporation of FA2 in the AAMs resulted in a significant drop in the thixotropic index values. For 50% fly ash addition in AAMs, it was observed that the drop of the thixotropic index was more noticeable in the case of FA2 relative to FA1, which is directly related to raw material properties, as will be discussed in the upcoming section.

As shown in Fig. 11b, the characteristic time results showed a similar trend to that of the thixotropic index. Relative to the FA-free pastes (0FA/100S-10%), the addition of FA1 caused an increase in the characteristic time values up to 50% FA1 content; however, the characteristic time dropped below that of the FA-free pastes value when using 55% FA1. Since the FA2 AAMs showed low thixotropic indexes, their corresponding characteristic times were not calculated.

5. Discussion

5.1 Effect of solid activator content on the rheology of one-part AAMs

The increase of solid activator content could possibly contribute both physically and chemically to higher thixotropy in the one-part AAMs. From a physical point of view, the use of higher activator content while fixing water usage increases the solid volume fraction in the system. Compared with two-part AAMs, the dissolution of solid activator proceeds gradually in the one-part AAMs. According to the ionic equilibrium, the rate of such a dissolution process slows down when the system becomes more saturated. In Fig. 12, a test showed that compared with 1-minute mixing, the one-part AAMs showed lower equilibrium shear stress when mixed for 10 minutes. It could be reasoned that further dissolution of solid activator after relatively longer mixing time decreases the equilibrium shear stress. Thus, in this study, the increase of solid activator content from 4% to 10% increases the volume of undissolved solid particles. Consequently, it induces an increase in static yield stress and thixotropy.

From a chemical perspective, the use of a relatively small activator content (4% to 6%) will provide negatively charged silicate species in the aqueous system that will create some repulsive forces between precursor grains. A previous study compared the rheology of GGBS/water paste and GGBS/sodium-silicate-solution paste and found that the GGBS activated by sodium silicate solution was more flowable than non-activated GGBS in water (Kashani et al. 2014) due to the repulsive forces initiated by the silicate species. On the other hand, the higher activator contents (8% to 10%) will elevate the pH value of the system and consequently prompts further dissolution of slag particles (more Ca2+ and Mg2+ cations in the system). The Ca2+/Mg2+ cations have double charges; thus, such cations may bridge precursors particles through silicate species or/and silanol groups, causing agglomeration. In this case, some attractive forces might develop among the precursor grains, which induces the thixotropic behavior of the one-part AAMs. The zeta potential results (Fig. 13) show that the increase of activator content resulted in...
more negative zeta values due to the higher silicate species content, reflecting the magnitude of electric double-layer forces between grains (Kashani et al. 2014).

5.2 Effect of fly ash type and content on the rheology of one-part AAMs

In the fresh state AAM, the FA will have two main contributions. Firstly, the spherical-shaped FA particles will induce a ball-bearing effect among the GGBS grains. Secondly, since the dissolution of FA is relatively slower compared to GGBS grains (Ismail et al. 2014; Qian et al. 2020), the FA will present a limited chemical contribution during the early stage reaction of one-part AAMs because the FA does not react with sodium silicates at ambient temperature (Palacios et al. 2019). However, it will dilute the active GGBS particles that are prone to dissolve under the high alkali environment and react with soluble silicate species. Previous studies (Panda et al. 2018; G et al. 2019) found that the addition of FA would reduce the yield stress of slag-based AAMs. However, in this study, FA1 increased the yield stress of one-part AAMs, which might be related to its physical properties, especially the carbon impurities, that possess a large specific surface area besides their irregular shape and porous nature. To draw attention to the influence of impurities on the rheology of FA/GGBS AAMs, FA1 was incinerated at 500°C for 1 hour, aiming to eliminate part of its available carbonaceous impurities. Figure 14 compares the stress decay response of the one-part AAMs using 50% GGBS along with 50% FA1 before and after incineration. Both initial and equilibrium stresses dropped, the thixotropic index decreased, and the characteristic time increased after incinerating FA1 at 500°C relative to the control FA1. This highlights the role of impurities and their contribution to the thixotropy of one-part AAMs. On the other hand, the yield stress of one-part AAMs significantly dropped with the addition of FA2, owing to its lower content of carbonaceous matters and lower specific surface area relative to FA1 (refer to Table 1).

6. Conclusions

The solid activator content significantly affects the rheology and thixotropy of the one-part AAMs. Both dynamic and static yield stresses, together with the thixotropic index, increase with increasing solid activator content. The slow dissolution of solid activator and electrostatic forces, as well as attraction forces due to Ca²⁺/Mg²⁺ cations, might be affecting the rheological and thixotropic properties of one-part geopolymer.

The type and content of fly ash also affect the rheological properties of blended one-part AAMs. The irregular-shaped impurities in FA are found to have a large surface area that absorbs water and consequently enhances thixotropy. Incinerating such type of FA at 500°C for 1 hour eliminates part of the carbonaceous impurities, and thus, their corresponding one-part AAMs achieve lower yield stress. On the contrary, FA with low carbon impurities decreases the static yield stress and thixotropy.

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