

Research Article

Durability and Microstructure Properties of Concrete with Arabic Gum Biopolymer Admixture

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Arabic gum biopolymer (AGB) has recently been demonstrated to improve mechanical and physical properties of fresh and hardened concrete which makes it a promising sustainable and environmentally friendly water-reducing admixture. The present work focuses on the effect of added AGB on the workability, setting time, and durability of concrete. Furthermore, a microstructure analysis is conducted to provide objective evidence and support for previous findings and hypothetic interpretations. Flow table experiments are conducted on Portland cement mortar mixed with different percentages of AGB to evaluate the workability. The initial setting time of cement paste is measured for different AGB contents. X-ray fluorescence tests are performed on cement-AGB mix powder to determine its chemical composition. Carbonation depth in AGB concrete samples is estimated to assess durability. AGB-added cement powder was subjected to X-ray diffraction and SEM tests to determine the rate of hydration and to expose the microstructure properties of AGB cement mix and help explain its macroscopic behavior, respectively.

1. Introduction

Arabic gum biopolymer (AGB) is a sustainable material extracted from wild plants. It is found mainly in Sudan [1] and in its neighborhood. It is known for its low viscosity and high solubility in water [2]. Owing to these attractive properties, it is employed in various industries including cosmetics, textiles, pharmaceuticals, encapsulation, lithography, and even the food industry. AGB has also been used as an additive in a binder to ceramic glazes to reduce the risk of damage during handling in the factory [3].

On the other hand, creating market opportunities for AGB benefits the local communities in producing countries through employment in the collection, extraction, and distribution of AGB.

Gum Acacia Karroo (GAK) comes from Acacia karroo Hayne, which grows mainly in the southern countries of Africa. GAK is a high molecular weight polysaccharide with a $M_w$ of $2.99 \times 10^6$ g-mol$^{-1}$ [4]. Arabic gum biopolymer is mostly referred to as gum from Acacia senegal and Acacia seyal and is mostly sought because of its purity and high grade [5]. AGB consists of a lower molecular weight polysaccharide with a $M_w$ of $0.25 \times 10^6$ g-mol$^{-1}$ [6, 7].

Both types of gums (AGB and GAK) contain percentages of natural sugars. They are made up of side chains of L-arabinose with D-glucuronic terminal unit or L-rhamnose with a backbone made up of D-galactose units [8].

In the earliest reported work [9] on the use of natural gum as an admixture for concrete, the study concluded that natural gum material improved workability and compressive strength.

In a subsequent work [4] the water to cement ratio (w/c) could be reduced from 0.61 to 0.48 and the compressive strength increased by 37% at 180 days of curing with the addition of Gum Arabic Karroo. With more added Gum Arabic Karroo, the density of concrete decreased slightly, the
workability improved, and a higher slump of 200% was obtained.

A mere visual observation [10] revealed a powerful effect of AGB against capillary diffusion, porosity, and capillarity. These properties were found to vary consistently with compressive strength and exhibited a minimum where strength was highest. Air content drops to a minimum as AGB fraction is increased. Workability was seen to increase with AGB content, which suggests a potential for the use of AGB in self-compacting concrete [10].

Flow test, bleeding, and compressive strength on cement mortar mixed with Arabic Gum Karroo as a water admixture were also investigated. The final setting time was increased by 6 h. Bleeding of mortar was decreased with the increase of Gum Acacia Karroo percentage. In the modified mixture, the lowest amount of entrained air was for a Gum Acacia Karroo percentage between 0.7% and 0.8%. The compressive strength was increased when the water to cement ratio could be reduced from 0.5 to 0.4 owing to a dosage of Gum Acacia Karroo between 0.7% and 0.9% [11].

In a more recent study [12], it was reported that the Arabic gum concrete (or biopolymer concrete) provides longer setting times and more workability than ordinary concrete.

A reduction (16%) in concrete permeability was reported in [13] for an optimal AGB weight fraction, which implies a significant improvement in durability. An improvement near 8% in flexural and tensile strength was obtained for an optimum proportion of AGB between 0.7% and 0.8%.

The aims of the present study are to, first, complement the investigation by studying more properties of concrete that are positively affected by the addition of AGB, namely, workability, density, initial setting time, and durability (carbonation) and, second, to analyze the chemical composition and microstructure of AGB concrete mixes (X-ray fluorescence) through X-ray diffraction and scanning electron microscopy (SEM) to support and confirm the interpretation of their observed macroscopic behavior.

2. Materials

2.1. Concrete. As a preliminary to the investigation of AGB concrete properties, a characterization of the ordinary Portland cement used in the study is carried out first.

The setting times of the cement were determined according to the Test Method for Time of Setting of Hydraulic Cement of the ASTM C191 Standard (2006) [14] using the Vicat apparatus. The specific gravity was obtained using the Test Method for Density of Hydraulic Cement of the ASTM C 188 Standard (2003) [15]. All these properties were evaluated using three data points per measured quantity. The results are shown in Table 1.

Then, the granulometry of the sand and the coarse aggregates was determined using sieving analysis according to the ASTM C 136-96a Standard Test Method (2007) [16] for Sieve Analysis of Fine and Coarse Aggregates. The obtained sizes for sand were 3/8, 8, 16, 30, 60, 100, and 200 μm. The coarse aggregates were between 1 inch and 3/8 grain size.

<table>
<thead>
<tr>
<th>Property name</th>
<th>Average value</th>
</tr>
</thead>
<tbody>
<tr>
<td>Initial setting time (min)</td>
<td>90</td>
</tr>
<tr>
<td>Final setting time (min)</td>
<td>190</td>
</tr>
<tr>
<td>Specific gravity</td>
<td>3.21</td>
</tr>
</tbody>
</table>

The fineness modulus of aggregate was defined as the weighted average size of sieves counted from the finest. The surface areas for aggregate samples have been calculated following [17] based on percent pass for each sieve. The properties of fine and coarse aggregates are presented in Table 2.

The sieve analysis results are presented in Figure 1. As shown, the obtained sample curve of passing percentage is located between the ASTM maximum and minimum curves. This means that the distributions of the aggregate particle properties such as size, percentage passing, and maximum weight on each sieve are in conformity with the standard.

2.2. Physicochemical Properties of AGB. Arabic gum biopolymer consists predominantly of volatile matter and a varying percentage of ash and contains metallic elements such as aluminum, iron, copper, zinc, and magnesium [1]. The physicochemical properties of AGB depend on the type and origin of the tree, exudation time, storage, and climate conditions. AGB is highly soluble in water and is characterized by a low viscosity compared to other gums. It can be dissolved in distilled water in concentrations beyond 50% [2]. Alkalinity of AGB is confirmed by a pure hydrogen (pH) of 8 measured for the AGB utilized in the present study using 781 pH/Ion Meter-Metrohm at a room temperature of 20.6°C.

2.3. Chemical Structural Unit of AGB. The chemical composition of AGB may depend on climate, origin, harvest season, age of tree, and processing condition.

AGB is a complex mixture consisting of polysaccharides and glycoproteins. It is the original source of the low sugars ribose and arabinose [18]. Recently, several investigations have been conducted in order to determine the molecular structure of AGB and relate it to its rheological and emulsifying properties.

The molecular structures of AGB is mainly formed by 3,6-chains linked β-D-galactopyranose substituted in position 6 by side chains of 3-linked α-L-arabinofuranose. Figure 2, presents the common structure of AGB [19].

The exact molecular structures of AGB are still rather uncertain because it is a mixture, and the material varies significantly depending on the source [20].

3. Methods

The flow table experiment was conducted to study the effect of AGB on the fluidity of mortar. The cone was placed on a clean and dry flow table, and the mold was filled with two layers of mortar that were each tamped 20 times using a
The tamping pressure was selected to ensure that the mold is filled with minimum void. The mold was then raised and removed from the mortar, and the flow was determined by dropping the table device 25 times in 15 seconds. The flow was calculated as the average of four measured diameters for four different directions and was expressed as a relative percentage of the original diameter. For each percentage of AGB, the retained result was based on the average of three samples. The experiment was carried out according to ASTM [21].

Initial setting time test was carried out for different dosages of AGB cement paste with a manual setting time apparatus (infraTest) with temperature between 20°C and 25°C. Six-hundred grams of OPC were mixed with different dosages of AGB between 0.1% and 1.1%. The 1 mm needle was released after 30 seconds, and the reading was taken just after mixing the paste. The second reading was taken after 30 minutes and the rest of readings every 15 minutes. The tests were performed according to ASTM C191-04 (2004) [22] as described in [23].

The density was evaluated as the ratio of the weight of the sample to the volume of the concrete mold as indicated per ASTM C642–13 (2013) [24]. The calculation was done for each specimen after curing to time of testing using an electronic balance with a sensitivity of 0.1 g.

PANalytical X-ray fluorescence (XRF) was applied to determine the chemical composition of both the AGB and the AGB cement mixes following 28 days of curing according to ASTM C114 as described in [25]. After crushing and passing a sieve diameter of 75 μm, the tested specimens were prepared in a Pt/Au crucible and then molded as pressed specimens. The powder is pressed into a solid tablet using a low-speed hydraulic press and a 40 mm die. The operation takes around 5 minutes per specimen.

The carbonation test was performed to evaluate the durability of the AGB concrete specimens. In the first step, cylindrical specimens with a diameter of 100 mm and height of 200 mm were prepared and subjected to curing for 28 days. Next, the surface of each sample was dried at laboratory temperature. Then, all specimens were placed in a carbonation chamber at a temperature of 30 ± 2°C and 61–69% room humidity. Moreover, the specimens were submerged in a 4% carbon dioxide (CO₂) solution for 28 days. Each sample was split through indirect tensile testing using a compression machine and directly sprayed with phenolphthalein. The reading of the carbonation penetration was taken at three points in each split, and then the average relative to the two split sides was determined. The procedure for the test was carried out following the RILEM committee CPC-18 as described in [26].

Samples of AGB and AGB cement were tested using XRD analysis to identify and compare their rates of hydration.

The powder method of XRD was carried out using Philips (PW 1730) with an X-ray source of Cu K-α radiation (with wave length \( \lambda = 1.5418 \) Å). The diffractometer was scanned for angles ranging from 5° to 60° with a step size of 0.015° at 1.8 s counting time per step. The current intensity and voltage of the X-ray device were fixed at 40 mA and 40 kV, respectively, as described in [27].

Using scanning electron microscopy (SEM), the field emission gun microscope with HV 15.00 kV, detector ETD, with addition of back-scattered electron detector, produces a strong electric field in order to stimulate electrons out of their atoms. It was used to determine the morphology of the hydration process produced in the specimen. An electronic gun at the top of the microscope directs vertically down a stream of electrons using a set of electromagnetic lenses inside a vacuum chamber.

The sample preparation manner has an important effect on the quality of the results. The tested specimens were prepared in powder form and then crushed and passed through sieve number 200. The specimen powder was fixed in around 1 cm diameter sample holder using a double side carbon tap and was coated in order to provide electrical conductivity in the powder specimen with a thin layer of gold using a sputter coater. For a complete drying, the samples were placed in a vacuum area inside the testing chamber inside the SEM.

### Table 2: Properties of fine and coarse aggregates.

<table>
<thead>
<tr>
<th>No</th>
<th>Property</th>
<th>Fine aggregate</th>
<th>Coarse aggregate</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Specific gravity</td>
<td>2.65</td>
<td>2.68</td>
</tr>
<tr>
<td>2</td>
<td>Water absorption (%)</td>
<td>0.62</td>
<td>0.40</td>
</tr>
<tr>
<td>3</td>
<td>Fineness modulus (%)</td>
<td>2.47</td>
<td>4.01</td>
</tr>
<tr>
<td>4</td>
<td>Total surface area (m²)</td>
<td>1.43</td>
<td>1.34</td>
</tr>
</tbody>
</table>

![Figure 1: Grain size distribution of fine and coarse aggregate.](image1)

![Figure 2: Molecular structures of AGB.](image2)
4. Results and Discussion

4.1. Flow Table Experiment. Flow test of AGB cement mortar at different dosage ranging from 0.1% to 1.1% has been determined as a horizontal spread measurement of AGB mortar mixes. Figure 3 presents the determined flow diameter of mortar as a function of AGB percentage. The effect of added AGB on the flow of mortar mixes is important. Relatively to the control specimen, the flow fluidity increases by 21% at 0.7% of AGB dosage and by 73% at 1.1%. This increased flow ability of AGB mortar mixes attributed to the high viscosity, rheological behavior, and emulsifying effect of AGB.

4.2. Setting Time. The samples were prepared for initial setting time tests for different percentages of AGB mixture. Results show that the measured initial setting time depends on the dosage of AGB percentage as presented in Figure 4. The initial setting time increases continuously with AGB dosage up to 0.9%. For the AGB percentage of 0.7%, the initial setting time was about 3.5 h. At 0.9%, it reached more than 8 h. The delay in initial setting time is useful in hot weather for early setting time. AGB can thus be used as retarding water reducer admixture in concrete mixes.

At a 0.4% of AGB dosage, the initial setting time was extended by more than 2.5 h compared to the control specimen. This provides better setting time than Pretoria gum that was only 1.8 h above the control [4].

In [28], retardation was found to depend on the tricalcium aluminate (C₃A) content in the cement. In [28], it is also mentioned that the retardation can be obtained when the calcium sulfate in the cement dissolves in water and reacts with C₃A. Thus, the addition of AGB in the water should activate a reaction of the tricalcium aluminate it contains with the calcium sulfate, which increases the setting time and then the retardation. This agrees with previous findings [4, 29] where some hydration products, together with C₃A, adsorb the gum which existed in the solution before the calcium sulfate reacted with C₃A.

The reasons for the extension of the setting time could be the decrease in the rate of ettringite production resulting from gypsum reaction with water and the hydration reaction that produces skeletal framework of calcium silicate hydrate (C₅S₈H₄) from alite (C₃S) and C₅S that react together. This agrees with the previous findings in [30].

4.3. Density of Concrete. The density of concrete sample corresponding to the explored dosages of AGB is presented in Figure 5. The sample density increases slightly with AGB proportions from 0% to 0.7%. At 0.9% of AGB, the density is slightly decreasing. This might occur due to the reduction of water cement ratio, which has a contribution in improving the strength by filling the microstructure pores of concrete. This pattern of variation in density closely follows that of compressive strength as reported in [13].

4.4. Chemical Composition. PANalytical X-ray fluorescence (XRF) was used to analyze the chemical composition of AGB-added cement after a specified period of curing. Table 3, presents some chemical components of AGB-added cement. Comparing the chemical composition before and after mixing of AGB, it is observed that some components, e.g., Al₂O₃, SiO₂, and Fe₂O₃, were reduced, whereas others such as SO₃ and CaO were increased.

It was reported in [31] that up to 4% weight fraction of CuO nanoparticles in cement could improve the mechanical and physical properties of concrete by reducing its porosity. This is in part achieved by increasing the crystalline of Ca(OH)₂ amount in the concrete structure.

The relative reduction of Al₂O₃ and TiO₂ fractions by 5% could contribute to the improvement in workability of fresh concrete given the indications in [32, 33] that the Al₂O₃ and TiO₂ nanoparticles, respectively, are favorable to the workability of fresh concrete.

The SiO₂ fraction’s relative decrease by 3% is among the factors that contribute to the prolongation of setting time according to Yazdi et al. [34] where that addition of SiO₂ nanoparticles is found to cause shortening in setting time.

The percentage of SO₃ was increased by 25%. SO₃ is reported in [35] to be responsible for the increase in cement mortar strength. Unlike other components which influence the strength properties of concrete through physical processes such as reduction of porosity, SO₃ does so by its chemical characteristics.
The study in [36] confirms that for an amount of TiO₂ up to 3.9%, the cement alite multiplies as the TiO₂ fraction goes down. However, a decrease by 2% only in TiO₂ fraction might not be enough to affect the growth of the alite in the cement. On the other hand, the 36% augmentation of BaO content is likely to cause the growth of the alite nanocrystals to greater dimensions as reported in [37].

It is reported in [38] that the addition of burnt pure CaO would increase the shear bond and the compressive strengths in specific conditions up to a 200°C temperature. CaO exhibited a modest increase of 0.3% that may not be significant enough to affect shear bond nor compressive strength. However, the 73% drop in Fe₂O₃ fraction may explain the observed improvement in the compressive and tensile strengths considering the results reported in [34] that indicate that up to 5% increase in Fe₂O₃ nanoparticle content reduces concrete strength.

4.5. Carbonation. Smaller carbonation depth in specimens indicates that the concrete has better carbonation resistances [39]. In Figure 6, it is shown that the depth of carbonation diminishes with the increase of AGB percentage. The carbonated part in the concrete specimen turned to purple due to the high alkalinity.

The decrease in the carbonation depth in AGB concrete specimen can be explained by the reduction in the water cement ratio and the ability of the AGB fine particles to occupy the microstructure voids which results in a more compact material. This is supported by the observed effect of AGB content on capillarity and porosity.

The obtained result for carbonation of concrete specimen agreed with the findings in [4] where it was indicated that the carbonation depth decreases with higher percentages of Arabic Gum Karroo.

4.6. X-Ray Diffraction Studies. XRD studies were carried out after 28 days of specimen curing time. The cement and the AGB cement samples with a dosage of 0.9% were analyzed. Figure 7 presents the different patterns of cement and AGB cement. The presented intensity patterns of the cement and AGB cement both are semicrystalline materials and almost have similar patterns. The chemical analysis on cement and AGB cement showed increases in some chemical components in the AGB mixes and provides new elements in concrete materials. This could improve the fresh and mechanical properties of concrete.

In Figure 7, a peak indicates the existence of a crystalline structure of the material, and the intensity of this peak
reflects the amount of this structure in the cement. For the AGB cement curve, it is observed that some intensities are higher (for examples for $2\theta$ around 22, 24, 29, and 39) than for the cement alone. It is also observed that some new peaks appear with small intensities. This agrees with the results of XRF test which indicated an increase in some crystalline structures such as Ca (OH)$_2$ at the early ages of hydration.

4.7. Microstructure Properties. Scanning electron microscopy (SEM) analysis was used to support the interpretation of the effect of AGB on concrete properties through examination of the void structure and microporosity [40]. The SEM results for cement and AGB cement are presented in Figures 8 and 9, respectively. For the control mix, Figure 8 shows the formation of CSH. The CSH was denser in AGB cement (Figure 9) than in the control specimen (Figure 8) due to the AGB emulsifying property that allows it to fill the voids in the cement microstructure and increase the material density. Finally, the spread of the initial CSH of the cement and the possible extra formed CSH produced by the AGB and cement reaction can be the origin of upgrading the concrete properties and providing better performance. This was attributed to the fact that the AGB concrete matrix was affected by the gum reaction, and then the Al (OH)$_2$ proportion was decreased which may reduce the porosity and permeability of the concrete and improve its durability.

The carbonation depths reported in Table 4 appear to increase moderately with GAK dosage whereas they drop notably with the addition of similar doses of AGB. This apparent contrast is explained by the difference in interpretation of the water dosages (w/c) used in the GAK and AGB cement mixes.

Additional results reported in [4] are based on the same mixing procedure used in the present work for AGB cement. They show that the carbonation depth increased slightly with the increase in GAK dosage, whereas with reduced w/c ratio the carbonation depth decreased and compressive strength increased by 3.7% and 37.03%, respectively, compared with control specimen [4]. This is because of the ability of the gum materials to replace the water and increase the density of concrete. Improvement in carbonation and compressive strength is in agreement the results obtained for the AGB cement mix.

5. Conclusion

Arabic gum biopolymer (AGB) is a vegetal product that is naturally grown in West Africa, most abundantly in Sudan. It is known for its numerous attractive physical and rheological properties. With the availability of AGB at low cost, it was tempting to investigate the impact of its properties on the performance of concrete when it is used as an admixture. In the present work, the effects of addition of AGB to cement mix on flow ability and on the initial setting time have been

![Figure 7: XRD patterns of AGB, cement, and AGB cement.](image7)

![Figure 8: SEM structure of pure cement material.](image8)

![Figure 9: SEM structure of AGB cement mixes.](image9)
Table 4: Properties of concrete with added biopolymer additives.

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<tbody>
<tr>
<td></td>
<td>Compressive strength (MPa) [10]</td>
<td>Tensile strength (MPa) [13]</td>
<td>Carbonation attack (mm)</td>
<td>Compressive strength (MPa)</td>
<td>Carbonation attack (mm)</td>
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<tr>
<td>0</td>
<td>43.8</td>
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</table>
studied experimentally. Series of tests have been conducted using concrete samples with various percentages of AGB to assess the influence of AGB on the density and carbonation depth of concrete and to determine the percentages that achieve the best performance for each property. X-ray fluorescence tests are performed to determine its chemical composition. Moreover X-ray diffraction and SEM tests were carried out to determine the microstructure properties of AGB cement mix. The test results showed that fluidity and initial setting time of the AGB mortar increase continuously with AGB content up to an optimal weight fraction of 0.9% AGB. The density slightly decreases with the increase of AGB dosage proportions from 0% to 0.9%. XRD results analysis showed almost similar hydration pattern of AGB cement relative to the OPC cement. The maximum relative improvement in the carbonation is around 11% with the increase of AGB percentage up to 0.9 percentage. The scanning electron microscopy (SEM) of AGB concrete shows less voids dispersed in the AGB mix microstructure. The use of AGB can be beneficial as a water-reducing admixture in the construction sector, resulting in decreasing the chemical admixture demand. Creating new outlets for the product would induce social and economic impacts.

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

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References


