Review

Waste Clay Bricks as a Geopolymer Binder for Pavement Construction

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Abstract: Geopolymer binders that combine aluminosilicate materials (i.e., precursors) with alkali activators are a viable and environmentally friendly alternative to ordinary Portland cement. While fly ash, slag, silica fume, and metakaolin are the most extensively investigated precursor materials, recent studies demonstrate the feasibility of using low amorphous aluminosilicates (LAA) for geopolymer synthesis. Waste clay bricks (WCB) make an excellent LAA material for producing geopolymer binders, considering their chemical and mineralogical properties. Geopolymer binders with enhanced mechanical properties can be produced either by blending WCB with other aluminosilicate materials or by using WCB as the sole precursor, while providing appropriate production conditions, such as high-temperature curing. Until now, in pavement construction, WCB has been investigated only as a subbase material or as an aggregate for concrete. Since WCB is a potential geopolymer source material, it can also function as an alternative cementitious material (ACM), and stabilizing material in pavement construction. This work reviews the recent studies on producing WCB-based geopolymers, with the focus particularly on the properties of raw materials, activator types and their concentrations, curing conditions, blended geopolymer systems, and the mechanical properties of WCB-based geopolymer binders. Simultaneously, different pavement design requirements and currently available specifications for the use of geopolymer concrete were correlated to evaluate their feasibility as an ACM in pavement construction. Based on the current literature, WCB can be proposed as a suitable ACM to develop pavement-grade concrete and more promising results can be obtained by blending WCB with high-calcium sources, such as slag. Therefore, comprehensive studies on geopolymer concrete development, durability, and field performance are recommended.

Keywords: waste clay bricks; geopolymer; activators; production conditions; compressive strength; rigid pavements

1. Introduction

Sealed transport pavements can be divided into two main categories: rigid pavements (often of concrete) and flexible pavements (or asphalt pavements) [1]. Concrete pavements have the advantage of high durability, low maintenance, low overall cost over the life cycle, and simple structure [2–5]. Generally, the maintenance cost of concrete pavements is 50% to 60% lower than that of asphalt pavements [5]. Furthermore, it is estimated that concrete pavements can be constructed almost 2.5 times faster than asphalt pavements, with 38% less equipment and 67% less labor [5]. Hence, concrete pavements have 18% to 28% less overall cost over their 30- to 40-year service life compared to any other pavement type [5–7]. As such, concrete pavements can be considered a viable and beneficial option to meet the future growth in transport demand.

Sustainability is becoming an increasingly central factor in modern transport pavement engineering. To ensure sustainability, all pavement performance conditions and standards must be achieved, while simultaneously reducing the associated environmental impacts...
by minimizing energy consumption, natural resource utilization, and emissions [8]. The primary challenge regarding sustainability in the concrete pavement industry is the high energy consumption and greenhouse gas emissions (i.e., CO$_2$) during ordinary Portland cement (OPC) manufacture [9–12]. The use of recycled, waste, and secondary materials to develop low-carbon concretes through geopolymer technology, which can then replace OPC-based concrete, is a common and successful strategy to enhance sustainability in the pavement industry [13].

Therefore, in the recent past, several studies were conducted on replacing Portland cement with suitable alternative cementitious materials (ACMs) [10,12,14]. These ACMs are usually materials that are rich in alumina and silica (i.e., aluminosilicate materials), such as fly ash, blast furnace slag, and silica fumes [15,16]. Combining these source materials with strong alkali solutions such as sodium hydroxide or potassium hydroxide “activates” them and creates geopolymer binders, which are a viable alternative to OPC [15–17].

There is a wide range of aluminosilicate materials that are generated as industrial by-products [18]. Almost all these materials have a relatively high percentage of alumina and silica in amorphous phases. Among them, fly ash, ground granulated blast-furnace slag, and silica fume are the most often used and extensively investigated materials [18–21]. These are abundantly available in some countries but can be extremely scarce in others. Therefore, searching for other suitable materials for geopolymers is always useful.

Interestingly, several studies on natural clay-based materials have found that even low amorphous precursors can produce a geopolymer of sufficient strength [22–27]. For instance, Molino et al. [24] investigated the use of river clay sediment to produce geopolymer binders. The raw material was calcined at 650 °C and 750 °C and then activated by potassium and sodium aluminate solutions at different concentrations. Similarly, Ferone et al. [23] evaluated reservoir clay as a geopolymer raw material after calcination at 650–750 °C and in combination with sodium hydroxide. Additionally, raw brick clay mixtures calcined at 750 °C were also found to produce geopolymer binders when mixed with alkali activators [25]. However, in all these cases, the high calcination temperatures required to process the raw materials are severely detrimental to both their sustainability indicators and cost [24,28].

On the other hand, clay obtained from waste bricks has the natural advantage of being already calcined at 850 °C to 950 °C in a kiln during the manufacturing process [29,30]. At these high calcination temperatures, the combined water in clay minerals evaporates, collapsing the crystalline clay network and creating disordered amorphous phases of silica and alumina [31]. With the presence of these phases, there is a high potential for waste clay bricks (WCB) to be an environmentally friendly and low-cost raw material for synthesizing geopolymers.

Of course, the local availability of the required raw materials is a vital factor for the viability of any large-scale field or commercial application [17]. Fortunately, a massive quantity of brick waste is generated every year through the brick manufacturing process [18] and from construction and demolition (C&D) waste [29,32,33]. Almost every country generates C&D waste, contributing almost 35% of the solid waste in the world [34,35]. For example, Europe generates more than 800 megatons of C&D waste per year [36,37], while the United States generates around 136 megatons per year [38]. Around 25% to 40% of the total waste generated in Australia is classified as C&D waste and approximately 17 megatons of waste masonry are generated annually, of which WCB is a major component [39,40]. When recycling C&D waste, priority is given to concrete waste, and the majority of WCB is still sent to landfills [40]. Reusing this waste for geopolymer binder development can reduce the carbon footprint in the construction industry while concurrently providing a profitable solution for solid waste management. Therefore, considering its availability, suitability, and additional environmental benefits, WCB is an economically and chemically viable candidate for producing geopolymer binders.

Studies by Fort et al. [28], Reig et al. [29], Tuyan et al. [18] and Silva et al. [22] investigated the feasibility of using WCB to synthesize geopolymer binders. These studies
reported WCB-based geopolymer binders with compressive strengths in the range of 30 MPa to 36 MPa. Several studies have been published investigating binary and ternary geopolymer systems, combining WCB with other aluminosilicate materials such as fly ash, slag, and metakaolin [30,41–43]. However, all these studies have employed a range of different production conditions such as curing conditions (i.e., curing temperature and curing time), the optimization of alkali activators, and the different sources of aluminosilicate materials. These factors can significantly influence the mechanical properties of the geopolymer matrix [22,32].

Initially, crushed WCB was only studied for its potential as fine and coarse aggregates in concrete production [44–49]. Specifically, for pavement construction, WCB was mainly investigated as a subbase construction material or as an aggregate in the base structure [50–56]. In contrast, the more recent studies by Migunthanna et al. [42,43] investigate the feasibility of using WCB as a binder in pavement geopolymer concretes. The purpose of this work is to present the current state-of-the-art in developing WCB-based geopolymers and to assess the feasibility of using WCB as a pavement construction material. The literature survey methodology is discussed in Section 2.

First, it is necessary to understand the design considerations, damage, and common failure types specific to rigid pavements. Hence, in Section 3, the minimum strength requirements according to different rigid pavement design guidelines (i.e., Australian standards and AASHTO standards), as well as the material properties affecting pavement failures are discussed. These are then correlated with the performance of WCB-based geopolymers, as recorded in the literature.

While a significant number of experimental and field studies are available on conventional geopolymer binders using fly ash, slag, silica fume, and metakaolin, only a few experimental studies report on geopolymers based on WCB precursors. However, they comprehensively discuss the different production conditions and the effect of those parameters on the performance of WCB-based binders. The scope of the research work reporting on WCB-based geopolymer synthesis and the different assessment criteria adopted for each study are summarized in Table 1. It is clear that none of the studies have evaluated the field performance of these low-carbon binders. Furthermore, the majority of the studies are limited to binder and mortar production, and they comprehensively evaluate the chemical and mechanical properties of WCB as a geopolymer binder. However, to the best of the authors’ knowledge, only the study by Migunthanna et al. [43] developed concrete using WCB-incorporated binders. Based on this literature, the chemical and mineralogical properties of WCB, collated from different sources, are discussed in Section 4, and the potential of WCB to serve as an aluminosilicate precursor is evaluated.

In Sections 5 and 6, the effects of alkali activator types and curing conditions on the mechanical properties of the resulting geopolymer are comprehensively reviewed. Summarizing the previous literature, conclusions are derived regarding suitable activator types, concentrations, and curing conditions optimal for WCB geopolymer synthesis. The possibility of further improving the performance of WCB geopolymers by combining WCB with other aluminosilicate materials is assessed in Section 7. The effect of the water-to-binder ratio, and the mineralogical and microstructural features of WCB-based geopolymer and its contribution to improving strength and durability, are reviewed in Sections 8 and 9, respectively.

Within the limited literature reported in this area, only a few studies have investigated and assessed the durability of WCB geopolymers in the long term. In Section 10, the findings of these studies on the structural integrity and durability of WCB-based geopolymers are summarized. Throughout this paper, the authors highlight the research gaps and future works necessary for developing WCB-based geopolymers that are suitable for rigid pavement applications.
Table 1. Summary of the scope of research work carried out related to WCB-based geopolymer synthesis: characterization techniques and functional properties.

<table>
<thead>
<tr>
<th>Reference</th>
<th>Characterization Techniques</th>
<th>Mechanical Properties</th>
<th>Durability Properties</th>
<th>Type of Study</th>
<th>Other Assessment Criteria</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>XRF</td>
<td>XRD</td>
<td>SEM</td>
<td>FTIR</td>
<td>Compressive Strength</td>
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<td>✓</td>
<td>✓</td>
<td>✓</td>
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<td>Silva et al. [22]</td>
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<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>7-day</td>
</tr>
<tr>
<td>Reig et al. [29]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>3, 7 days</td>
</tr>
<tr>
<td>Fort et al. [28]</td>
<td>✓</td>
<td>✓</td>
<td>-</td>
<td>-</td>
<td>28-day</td>
</tr>
<tr>
<td>Fort et al. [57]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Sassoni et al. [58]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>Tuyan et al. [18]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>1, 3, 5, 7, 28, 90 days</td>
</tr>
<tr>
<td>Rovnakik et al. [41]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>-</td>
<td>7, 28, 90 days</td>
</tr>
<tr>
<td>Rovnakik et al. [59]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>7, 14 days</td>
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<tr>
<td>Zawrah et al. [30]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>3, 7, 28, 90 days</td>
</tr>
<tr>
<td>Ouda and Gharieb [35]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>1, 7, 14, 28 days</td>
</tr>
<tr>
<td>Robayo-Salazar et al. [60]</td>
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<td>Vafaei and Allahverdi [62]</td>
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<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>28-day</td>
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<tr>
<td>Vafaei et al. [62]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>At different durations over 24 months after exposure to acid</td>
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<tr>
<td>Reference</td>
<td>Characterization Techniques</td>
<td>Mechanical Properties</td>
<td>Durability Properties</td>
<td>Type of Study</td>
<td>Other Assessment Criteria</td>
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<td>Mahmoodi et al. [63]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓ - 7, 28 days - -</td>
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<tr>
<td>Ulugol et al. [20]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓ - 24, 48, 72 h - -</td>
</tr>
<tr>
<td>Wong et al. [64]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓ - 1, 28 days - -</td>
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<tr>
<td>Migunthanna et al. [42]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓ - 1, 3, 5, 7, 14, 28, 60, 90 days - -</td>
</tr>
<tr>
<td>Migunthanna et al. [43]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓ - 3, 7, 28 days - -</td>
</tr>
<tr>
<td>Cong et al. [65]</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓</td>
<td>✓ - 7, 28 days - Thermogravimetric analysis - -</td>
</tr>
</tbody>
</table>
2. Review Methodology

Data from peer-reviewed journal articles were mainly used to evaluate the different chemical, mineralogical and microstructural properties, production conditions, mechanical performance, and durability of WCB-based geopolymer binders. Data from pavement design guidelines, test methods, and the currently available specifications for the use of geopolymer concrete were correlated to evaluate their feasibility as an ACM in pavement construction.

The related literature was searched in the Web of Science (WoS), Google Scholar, and Scopus databases. Bibliometric analysis was carried out in the WoS database using the search string (“Waste clay brick” OR “brick waste” OR “waste brick”) NOT aggregates AND geopolymer. A total number of 26 journal articles were extracted within the period from 2013 to 2022. Among these, almost 85% were published recently, in the last 5 years. This indicates the recent research interest in using WCB as a geopolymer precursor.

The network visualization and density visualization diagrams, based on keyword co-occurrence analysis for the above search, were obtained through VOSviewer version 1.6.17 bibliometric analysis software, as shown in Figure 1a,b, respectively. A total of 10 items, 2 clusters, and 42 links were identified in the network visualization. Among these, compressive strength, geopolymer, and microstructure were identified as the most intense keywords. Furthermore, when the search string was modified by adding “AND pavement”, the total journal articles found were reduced to 1, pointing to the limited research available on using WCB as a binder material in pavement construction.

![Network visualization and density visualization diagram](image)

Figure 1. (a) Network visualization and (b) density visualization diagram, based on keyword co-occurrence analysis corresponding to the WoS data.

3. Design of Concrete Pavements

The rigid pavement design guidelines followed in different countries and regions vary depending on their climate, geography, and geology. In this work, Australian pavement design guidelines are mainly considered. To design the base thickness of the pavement, 28-day concrete’s flexural strength is used as the design strength and is required to be
above 5 MPa [1]. Flexural strength can be approximated by Equation (1), as defined in the Austroads Guide to Pavement Technology Part 2: Pavement Structural Design [1]:

\[
f_{cf} = 0.75 \times \sqrt{f_c}
\]

where \( f_{cf} \) is the 28-day concrete’s flexural strength and \( f_c \) is the 28-day concrete’s compressive strength in MPa.

Accordingly, base concrete is expected to have a 28-day compressive strength of not less than 32 MPa [1]. These required minimum strengths can vary slightly, based on the design standards followed. For example, the minimum compressive strength defined in AASHTO guidelines is around 15 MPa, prior to opening to traffic, followed by 28 MPa after 28 days [66]. Considering the practical aspects of road projects, concretes with high early strengths are, hence, always recommended [67].

In addition to constructing the base layer, concrete is also used at the subbase layer in rigid pavements. This concrete type is known as lean-mixed concrete, which requires a minimum compressive strength of 5 MPa at 28 days [1]. To produce both these types and grades of concrete, OPC can often be replaced with geopolymer binders to obtain geopolymer concrete [68].

The majority of the damage and failures of rigid pavements occur because of self-deterioration, due to low-quality and non-durable materials, and because of poor construction [69]. Cracks are the most common type of concrete pavement deterioration, and they occur in many ways, as shown in Figure 2a, and for different reasons [9,69]. Material properties are crucial in increasing the pavement’s resistance to deterioration. The most important material properties of pavement concrete are its workability, durability, strength, stiffness, and dimensional stability when facing temperature and moisture variations [9]. The constructability of the pavement structure depends on the workability of the fresh mixture, in terms of its ease of placing and finishing [9]. Improving the dimensional stability of concrete reduces the excessive strains, stresses, and deflections that develop in the pavement structure under the combined effects of traffic loading and environmental conditions (i.e., temperature and moisture) [70]. If the concrete is dimensionally unstable when under stress (i.e., curling, wrapping, contraction, expansion, and shrinkage), it will result in damage, such as cracking, corner breaks, and blow-ups (Figure 2) [69,70]. Furthermore, to ensure the durability and performance of the structure, the concrete should have good resistance to freeze-thawing, aggressive chemicals, and abrasion [9,69].

Figure 2. Examples of common types of pavement deterioration: (a) cracking, (b) blowups.
4. Properties of WCB as a Binder

4.1. Chemical and Mineralogical Properties

The X-ray diffraction (XRD) data shows quartz (SiO$_2$) to be the major mineralogical phase in WCB, irrespective of the material source [18,22,29,30,33,35,41,57,59,61]. The X-ray fluorescence (XRF) data presented in Figure 3 further verifies the presence of silicon oxide (SiO$_2$) as the major compound of WCB. Muscovite (KAl$_2$(AlSi$_3$O$_{10}$)(OH)$_2$), albite (NaAlSi$_3$O$_8$), and hematite (Fe$_2$O$_3$) are the other commonly occurring minerals in WCB [18,22,29,30,33,35,41,57,59,61]. The addition of sand during the brick manufacturing process and the content of the raw clay determine the levels of these mineral phases in WCB [30].

A typical raw brick clay mixture is composed of kaolinite, muscovite, gypsum, carbonates, and quartz as its main mineralogical phases [25]. However, after calcination at $750^\circ$C, the kaolinite and gypsum phases disappear, while the quartz and muscovite remain stable [25]. In summary, the calcination of bricks during its manufacturing process makes quartz the dominant mineralogical phase of WCB.

In geopolymerization, the degree of reaction over time is governed by the amorphous content of the source material [71]. SiO$_2$ and aluminium oxide (Al$_2$O$_3$) are the main amorphous phases in WCB [58]. WCB as a precursor shows low amorphousness, due to the low cooling rate of bricks after calcination during the brick manufacturing process [18]. For example, Foít et al. [57] reported 28% of amorphous phase in WCB sourced from brick grinding. Rovnaník et al. [41,59] reported 22% and 18.49% amorphousness for WCB samples sourced from brick grinding and skiving processes, respectively. However, relative to other C&D wastes, such as concrete and glass waste, WCB shows significant potential for geopolymerization due to its high SiO$_2$ and Al$_2$O$_3$ contents [12,32].

The chemical composition of an ACM intended for use in concrete should record a total SiO$_2$, Al$_2$O$_3$, and iron oxide (Fe$_2$O$_3$) content of at least 70%, while also recording the presence of calcium oxide (CaO) [72,73]. WCB is shown to be capable of satisfying the above chemical requirements since it contains SiO$_2$, Al$_2$O$_3$, Fe$_2$O$_3$, and CaO as its major compounds. Figure 3 illustrates the XRF results for SiO$_2$, Al$_2$O$_3$, Fe$_2$O$_3$, and CaO percentages in WCB from different sources, as reported in ten different studies. Table 2 shows the different sources of raw WCB used for producing geopolymers. Reig et al. [29], Komnitas et al. [32], Robayo-Salazar et al. [33], Ouda and Gharieb [35] and Silva et al. [22] used WCB recovered from C&D waste, and they all satisfied the basic chemical requirements. In contrast, Guo et al. [61] and Rovnaník et al. [59] used WCB from the brick grinding process, and in both cases, the sum total of the SiO$_2$, Al$_2$O$_3$, and Fe$_2$O$_3$ contents was below 70%. This can be attributed to the nature of the clay and the sand content used in the brick manufacturing process.

In addition, Guo et al. [61] studied a nano-modified geopolymer produced by combining 30% WCB with 70% Class C fly ash. According to the composition of fly ash given by Guo et al. [61], the sum of SiO$_2$, Al$_2$O$_3$, and Fe$_2$O$_3$ in the final mixture (i.e., WCB with fly ash) is 73.58%. Similarly, Rovnaník et al. [59] substituted 0, 25, 50, 75, and 100% of the WCB with metakaolin. Except for the mixture with only WCB, all other mixtures satisfied the SiO$_2$, Al$_2$O$_3$, and Fe$_2$O$_3$ composition requirements.
Table 2. WCB-based geopolymer material properties, reported in the literature: raw WCB material details, activators, curing conditions, and compressive strengths.

(* Note: $d_{avg}$ = average particle size, $d_{50}$ = median particle size, $d_{max}$ = maximum particle size, n.a. = not available).

<table>
<thead>
<tr>
<th>Reference</th>
<th>Compressive Strength (MPa)</th>
<th>Days</th>
<th>Curing Temperature (°C)</th>
<th>Relative Humidity (%)</th>
<th>Activator Type</th>
<th>Activator Content (by wt% of Binder)</th>
<th>Liquid/Binder Ratio</th>
<th>Particle Size</th>
<th>Source of Brick Waste</th>
</tr>
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<tr>
<td>Komnitsas et al. [32]</td>
<td>49.5</td>
<td>7</td>
<td>90</td>
<td>-</td>
<td>NaOH + Na$_2$SiO$_3$</td>
<td>NaOH: 8M, 5% Na$_2$SiO$_3$: Ms 3.5, 6%</td>
<td>0.38</td>
<td>$d_{avg}$ = 140 µm, $d_{50}$ = 6.6 µm</td>
<td>C&amp;D waste</td>
</tr>
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<td>Silva et al. [22]</td>
<td>37 ± 3</td>
<td>7</td>
<td>80</td>
<td>-</td>
<td>NaOH + Na$_2$SiO$_3$</td>
<td>act Na$_2$O = 8% (act Ms = 0.6)</td>
<td>0.27</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = 19.66 µm</td>
<td>C&amp;D waste</td>
</tr>
<tr>
<td>Reig et al. [29]</td>
<td>14.75 ± 0.25</td>
<td>7</td>
<td>65</td>
<td>-</td>
<td>NaOH</td>
<td>act Na$_2$O = 7% (act Ms = 1.6)</td>
<td>0.45</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = 20.9 µm</td>
<td>C&amp;D waste</td>
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<tr>
<td></td>
<td>29 ± 1</td>
<td>7</td>
<td>65</td>
<td>-</td>
<td>NaOH + Na$_2$SiO$_3$</td>
<td>act Na$_2$O = 7% (act Ms = 2.0)</td>
<td>0.30</td>
<td></td>
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</tr>
<tr>
<td></td>
<td>50 ± 2.5</td>
<td>7</td>
<td>65</td>
<td>-</td>
<td>NaOH + Na$_2$SiO$_3$</td>
<td>act Na$_2$O = 7% (act Ms = 2.0)</td>
<td>0.30</td>
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<td>41.9</td>
<td>28</td>
<td>20</td>
<td>-</td>
<td>NaOH + Na$_2$SiO$_3$</td>
<td>NaOH: 1.3% Na$_2$SiO$_3$: 33%</td>
<td>0.57</td>
<td>$d_{avg}$ = 125 µm, $d_{50}$ = n.a.</td>
<td>Waste from brick grinding</td>
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<td>Not assessed</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>NaOH + Na$_2$SiO$_3$ + NaAlO$_2$</td>
<td>-</td>
<td>n.a</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = 10 µm</td>
<td>Bricks with physical and geometric defects</td>
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<tr>
<td>Fort et al. [57]</td>
<td>Not assessed</td>
<td>-</td>
<td>60, 80</td>
<td>50</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 0.8)</td>
<td>act Na$_2$O = 7%</td>
<td>-</td>
<td>$d_{avg}$ = 125 µm, $d_{50}$ = 50 µm</td>
<td>Waste from brick grinding</td>
</tr>
<tr>
<td>Tuyan et al. [18]</td>
<td>36.2</td>
<td>5</td>
<td>90</td>
<td>80</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.6)</td>
<td>act Na$_2$O = 10%</td>
<td>0.46</td>
<td>$d_{avg}$ = 100 µm, $d_{50}$ = n.a.</td>
<td>Bricks damaged during production</td>
</tr>
<tr>
<td></td>
<td>34.9</td>
<td>7</td>
<td>25 ± 2</td>
<td>40</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.6)</td>
<td>act Na$_2$O = 10%</td>
<td>0.46</td>
<td>$d_{avg}$ = 100 µm, $d_{50}$ = n.a.</td>
<td>Bricks damaged during production</td>
</tr>
<tr>
<td></td>
<td>0</td>
<td>5</td>
<td>21 ± 1</td>
<td>80</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.6)</td>
<td>act Na$_2$O = 10%</td>
<td>0.46</td>
<td>$d_{avg}$ = 100 µm, $d_{50}$ = n.a.</td>
<td>Bricks damaged during production</td>
</tr>
<tr>
<td></td>
<td>0.5</td>
<td>3</td>
<td>28</td>
<td>21 ± 2</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.0)</td>
<td>25%</td>
<td>0.38</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = n.a.</td>
<td>Waste from brick skiving</td>
</tr>
<tr>
<td></td>
<td>5</td>
<td>7</td>
<td>29</td>
<td>50 ± 2</td>
<td>NaOH + Na$_2$SiO$_3$ (vol. Na$_2$SiO$_3$/NaOH = 2.5)</td>
<td>30%</td>
<td>0.30</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = 1.1 µm</td>
<td>Waste produced during brick manufacturing</td>
</tr>
<tr>
<td></td>
<td>10</td>
<td>28</td>
<td>90</td>
<td></td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.4)</td>
<td>-</td>
<td>-</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = 8.5 µm</td>
<td>Waste from brick grinding</td>
</tr>
<tr>
<td>Rovnanik et al. [41]</td>
<td>8 ± 0.5</td>
<td>7</td>
<td>28</td>
<td>21 ± 2</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.0)</td>
<td>25%</td>
<td>0.38</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = n.a.</td>
<td>Waste from brick skiving</td>
</tr>
<tr>
<td></td>
<td>21 ± 1</td>
<td>28</td>
<td>90</td>
<td>50 ± 2</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.0)</td>
<td>25%</td>
<td>0.38</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = n.a.</td>
<td>Waste from brick skiving</td>
</tr>
<tr>
<td></td>
<td>41 ± 5</td>
<td>28</td>
<td>90</td>
<td>50 ± 2</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.0)</td>
<td>25%</td>
<td>0.38</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = n.a.</td>
<td>Waste from brick skiving</td>
</tr>
<tr>
<td></td>
<td>~3</td>
<td>3</td>
<td>28</td>
<td>21 ± 2</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.0)</td>
<td>25%</td>
<td>0.38</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = n.a.</td>
<td>Waste from brick skiving</td>
</tr>
<tr>
<td></td>
<td>~5</td>
<td>7</td>
<td>28</td>
<td>50 ± 2</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.0)</td>
<td>25%</td>
<td>0.38</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = n.a.</td>
<td>Waste from brick skiving</td>
</tr>
<tr>
<td></td>
<td>~10</td>
<td>28</td>
<td>90</td>
<td>50 ± 2</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.0)</td>
<td>25%</td>
<td>0.38</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = n.a.</td>
<td>Waste from brick skiving</td>
</tr>
<tr>
<td>Zawrah et al. [30]</td>
<td>~3</td>
<td>3</td>
<td>28</td>
<td>50 ± 2</td>
<td>NaOH + Na$_2$SiO$_3$ (vol. Na$_2$SiO$_3$/NaOH = 2.5)</td>
<td>30%</td>
<td>0.30</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = 1.1 µm</td>
<td>Waste produced during brick manufacturing</td>
</tr>
<tr>
<td>Rovnanik et al. [59]</td>
<td>7 ± 0.75</td>
<td>7</td>
<td>28</td>
<td>80 °C for 24 h, after, at ambient temperature</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.4)</td>
<td>-</td>
<td>-</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = 8.5 µm</td>
<td>Waste from brick grinding</td>
</tr>
<tr>
<td></td>
<td>~15</td>
<td>7</td>
<td>28</td>
<td>80 °C for 24 h, after, at ambient temperature</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.4)</td>
<td>-</td>
<td>-</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = 8.5 µm</td>
<td>Waste from brick grinding</td>
</tr>
<tr>
<td></td>
<td>7.49</td>
<td>28</td>
<td>23</td>
<td>25 °C</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.4)</td>
<td>-</td>
<td>-</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = 8.5 µm</td>
<td>Waste from brick grinding</td>
</tr>
<tr>
<td></td>
<td>11.24</td>
<td>28</td>
<td>23</td>
<td>70 °C for 24 h, after, at 25 °C</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.4)</td>
<td>-</td>
<td>-</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = 8.5 µm</td>
<td>Waste from brick grinding</td>
</tr>
<tr>
<td></td>
<td>54.38</td>
<td>28</td>
<td>23</td>
<td>70 °C for 24 h, after, at 25 °C</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.4)</td>
<td>-</td>
<td>-</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = 8.5 µm</td>
<td>Waste from brick grinding</td>
</tr>
<tr>
<td></td>
<td>66.56</td>
<td>28</td>
<td>23</td>
<td>70 °C for 24 h, after, at 25 °C</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.4)</td>
<td>-</td>
<td>-</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = 8.5 µm</td>
<td>Waste from brick grinding</td>
</tr>
<tr>
<td></td>
<td>7.49</td>
<td>28</td>
<td>23</td>
<td>70 °C for 24 h, after, at 25 °C</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.4)</td>
<td>-</td>
<td>-</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = 8.5 µm</td>
<td>Waste from brick grinding</td>
</tr>
<tr>
<td></td>
<td>11.24</td>
<td>28</td>
<td>23</td>
<td>70 °C for 24 h, after, at 25 °C</td>
<td>NaOH + Na$_2$SiO$_3$ (act Ms = 1.4)</td>
<td>-</td>
<td>-</td>
<td>$d_{avg}$ = n.a., $d_{50}$ = 8.5 µm</td>
<td>Waste from brick grinding</td>
</tr>
</tbody>
</table>
Table 2. Cont.

<table>
<thead>
<tr>
<th>Reference</th>
<th>Compressive Strength (MPa)</th>
<th>Days</th>
<th>Curing Temperature (°C)</th>
<th>Relative Humidity (%)</th>
<th>Activator Type</th>
<th>Activator Content (by wt% of Binder)</th>
<th>Liquid/Binder Ratio</th>
<th>Particle Size</th>
<th>Source of Brick Waste</th>
</tr>
</thead>
<tbody>
<tr>
<td>Robayo-Salazar et al. [60]</td>
<td>2 ± 0.5</td>
<td>7</td>
<td>25 ± 3</td>
<td>80%</td>
<td>NaOH</td>
<td>10%</td>
<td>0.23</td>
<td>d&lt;sub&gt;avg&lt;/sub&gt; = n.a.</td>
<td>Rejected samples from the brick industry</td>
</tr>
<tr>
<td></td>
<td>7 ± 0.5</td>
<td>28</td>
<td>25 ± 3</td>
<td>80%</td>
<td>NaOH + Na&lt;sub&gt;2&lt;/sub&gt;SiO&lt;sub&gt;3&lt;/sub&gt;</td>
<td>44%</td>
<td>0.23</td>
<td>d&lt;sub&gt;avg&lt;/sub&gt; = 24.25 µm</td>
<td></td>
</tr>
<tr>
<td></td>
<td>5 ± 0.5</td>
<td>7</td>
<td>25 ± 3</td>
<td>80%</td>
<td>NaOH + Na&lt;sub&gt;2&lt;/sub&gt;SiO&lt;sub&gt;3&lt;/sub&gt;</td>
<td>44%</td>
<td>0.23</td>
<td>d&lt;sub&gt;avg&lt;/sub&gt; = 24.25 µm</td>
<td></td>
</tr>
<tr>
<td></td>
<td>60 ± 2</td>
<td>28</td>
<td>25 ± 3</td>
<td>80%</td>
<td>NaOH + Na&lt;sub&gt;2&lt;/sub&gt;SiO&lt;sub&gt;3&lt;/sub&gt;</td>
<td>44%</td>
<td>0.23</td>
<td>d&lt;sub&gt;avg&lt;/sub&gt; = 24.25 µm</td>
<td></td>
</tr>
<tr>
<td>Migunthanna et al. [42]</td>
<td>5.4</td>
<td>28</td>
<td>25 ± 3</td>
<td>-</td>
<td>Na&lt;sub&gt;2&lt;/sub&gt;SiO&lt;sub&gt;3&lt;/sub&gt;</td>
<td>10%</td>
<td>0.32</td>
<td>d&lt;sub&gt;avg&lt;/sub&gt; = n.a.</td>
<td>C&amp;D waste</td>
</tr>
<tr>
<td></td>
<td>10.5</td>
<td>90</td>
<td>25 ± 3</td>
<td>-</td>
<td>Na&lt;sub&gt;2&lt;/sub&gt;SiO&lt;sub&gt;3&lt;/sub&gt; (act Ms = 1, solid form)</td>
<td>10%</td>
<td>0.32</td>
<td>d&lt;sub&gt;avg&lt;/sub&gt; = n.a.</td>
<td>C&amp;D waste</td>
</tr>
<tr>
<td></td>
<td>~2</td>
<td>28</td>
<td>25 ± 3</td>
<td>-</td>
<td>Na&lt;sub&gt;2&lt;/sub&gt;SiO&lt;sub&gt;3&lt;/sub&gt; (act Ms = 1, solid form)</td>
<td>10%</td>
<td>0.32</td>
<td>d&lt;sub&gt;avg&lt;/sub&gt; = n.a.</td>
<td>C&amp;D waste</td>
</tr>
<tr>
<td></td>
<td>~3.5</td>
<td>90</td>
<td>25 ± 3</td>
<td>-</td>
<td>Na&lt;sub&gt;2&lt;/sub&gt;SiO&lt;sub&gt;3&lt;/sub&gt; (act Ms = 1, solid form)</td>
<td>10%</td>
<td>0.32</td>
<td>d&lt;sub&gt;avg&lt;/sub&gt; = n.a.</td>
<td>C&amp;D waste</td>
</tr>
<tr>
<td>Ulugol et al. [20]</td>
<td>~42</td>
<td>2</td>
<td>95</td>
<td>-</td>
<td>NaOH</td>
<td>10%</td>
<td>0.35</td>
<td>d&lt;sub&gt;50&lt;/sub&gt; = 7.6 µm</td>
<td>C&amp;D waste</td>
</tr>
<tr>
<td></td>
<td>~45</td>
<td>3</td>
<td>105</td>
<td>-</td>
<td>NaOH</td>
<td>12%</td>
<td>0.35</td>
<td>d&lt;sub&gt;50&lt;/sub&gt; = 46.9 µm</td>
<td>C&amp;D waste</td>
</tr>
<tr>
<td></td>
<td>~43</td>
<td>2</td>
<td>115</td>
<td>-</td>
<td>NaOH</td>
<td>15%</td>
<td>0.35</td>
<td>d&lt;sub&gt;50&lt;/sub&gt; = 46.9 µm</td>
<td>C&amp;D waste</td>
</tr>
</tbody>
</table>
4.2. Particle Size Distribution

The particle size distribution of the raw materials significantly affects the mechanical properties of WCB-based geopolymers [32]. Table 2 shows the particle size of the raw WCB used in different studies. The finer raw materials provide a higher surface area for the geopolymerization reaction [12,28,32]. Consequently, the aluminosilicate network becomes denser and stronger, resulting in better strengths [12,28]. Microscopic observations show that the larger particles react only partially during geopolymerization, causing a heterogeneous microstructure [29,30]. In other words, smaller particles react and dissolve completely during the geopolymerization, while larger particles remain partially unreacted [30]. The presence of these unreacted or partially reacted products reduce the compressive strength [29]. This mechanism is further evidenced by the decrease in compressive strength from 35 MPa to 9 MPa when the particle size was increased from 140 µm to 350 µm [32].

5. Effect of the Types and Concentrations of Activators in WCB-Based Geopolymers

5.1. Methods of Mixing Activators

When preparing geopolymer binders, activators can be used, either in aqueous or in solid form. Conventional geopolymers are produced by combining aluminosilicate source materials with alkali activators in an aqueous form [74,75]. Since this method consists of adding two liquid parts (i.e., water and activators), these geopolymers are referred to as two-part geopolymers [75]. In contrast, one-part geopolymers are produced by adding solid alkali activators to obtain a dry mixture; then, similar to OPC, the reaction will start after adding water [17,75,76]. According to the currently reported studies, sodium silicate (i.e., water glass, Na₂SiO₃) and sodium hydroxide (NaOH) were the most frequently used activators to produce a two-part geopolymer. Only the more recent studies published by Migunthanna et al. [42,43] have reported synthesizing one-part WCB-based geopolymer.

5.2. Types of Activators

The type and content of alkali activators play an important role in the geopolymerization process [77]. Table 2 shows the different types of activators used in WCB geopolymer production. NaOH and combined NaOH-Na₂SiO₃ are the most commonly used activators. Only the study by Robayo-Salazar et al. [60] reported using sodium sulfate (Na₂SO₄) and sodium carbonate (Na₂CO₃) activators. This study recorded maximum compressive strengths of around 33 MPa and 21 MPa for Na₂SO₄ and Na₂CO₃, respectively. Figure 4 illustrates the 28-day compressive strength of WCB- and OPC-based geopolymers obtained by Robayo-Salazar et al. [60] for different types of alkali activators, with the same percentage of OPC replacement by WCB. Since these geopolymer precursors consist of 80% WCB and 20% OPC, a direct comparison of the effect of Na₂SO₄ and Na₂CO₃ activators on 100% WCB-based geopolymer is not possible.
Figure 3. Content of SiO$_2$, Al$_2$O$_3$, Fe$_2$O$_3$, and CaO in raw WCB, as recorded in the [22,28–32,33,35,41,59,61].

Figure 4. The 28-day compressive strength of the combined WCB and OPC precursors (80% WCB and 20% OPC) activated with different alkali activators. (source of data: Robayo-Salazar et al. [60] (Figure 5)).
Figure 5. Comparison of the compressive strengths of WCB geopolymers produced with NaOH, combined NaOH–Na$_2$SiO$_3$, and Na$_2$SiO$_3$ activators. ($t =$ curing temperature, $l/b =$ liquid-to-binder ratio, $w/b =$ water-to-binder ratio) [29,42,60].

Na$_2$SO$_4$ and Na$_2$CO$_3$ show a poorer activation potential of WCB [60] compared to NaOH and the combined NaOH–Na$_2$SiO$_3$. Furthermore, using Na$_2$SiO$_3$ in the alkali activation of WCB results in significantly higher compressive strengths compared to NaOH alone [60]. This is because the soluble silica present in Na$_2$SiO$_3$ encourages the formation of geopolymerization products, while also providing a strong interface between the unreacted particles and the geopolymer matrix [33,60,78,79].

Figure 5 shows a comparison of the compressive strength of WCB geopolymer produced with NaOH, Na$_2$SiO$_3$, and combined NaOH–Na$_2$SiO$_3$ activators across three different studies. At an ambient curing temperature, Robayo-Salazar et al. [60] obtained maximum 7-day compressive strengths of around 2 MPa and 5 MPa for NaOH and combined NaOH–Na$_2$SiO$_3$ activators, respectively. Similarly, Reig et al. [29] recorded 7-day compressive strengths of around 15 MPa and 29 MPa for WCB activated with NaOH and combined NaOH–Na$_2$SiO$_3$, respectively (for both samples, curing temperature = 65 °C, liquid/binder = 0.45). Furthermore, at an ambient curing temperature, Robayo-Salazar et al. [60] obtained a maximum 28-day compressive strength of 54.38 MPa for WCB activated with combined NaOH–Na$_2$SiO$_3$, which is 86% higher than the strength of WCB activated with NaOH alone. These examples show that irrespective of the curing temperature, the addition of Na$_2$SiO$_3$ to the activator can result in better compressive strengths. The addition of soluble silicates to the activator can accelerate the polycondensation process when compared to using alkaline hydroxides alone [12,64].

Since NaOH is a highly corrosive, viscous, and hazardous chemical, the construction industry shows a reluctance to use geopolymer binders in commercial applications [17,75,80]. Therefore, discounting NaOH, it is important to evaluate the effect of Na$_2$SiO$_3$ as the sole activator in producing WCB-based geopolymers. The studies by Migunthanna et al. [42,43] synthesized WCB-based one-part geopolymer binders, using Na$_2$SiO$_3$ as the sole activator. Under ambient curing, their one-part WCB binders achieved a 7-day compressive strength of 3.3 MPa, which is higher than the strength achieved with only a NaOH activator. However, this is still less than for the combined NaOH–Na$_2$SiO$_3$ activator (Figure 5). Hence, all the current literature agrees that the best mechanical performance for a 100% WCB-based geopolymer is obtained when using a combined NaOH–Na$_2$SiO$_3$ activator. However, Na$_2$SiO$_3$, as the sole activator, showed a similar mechanical performance to a combined...
NaOH–Na$_2$SiO$_3$ activator for blends of WCB with other aluminosilicate precursors, such as fly ash and slag [42,43].

5.3. Concentration of Activators

The silica modulus (Ms) of the activator is an important parameter in characterizing the alkali activator and is defined as the molar ratio between SiO$_2$ and Na$_2$O. Ms determines the amount of soluble silicates present and is, therefore, a critical factor in controlling the dissolution rate and gelation that occurs during the geopolymerization process [16]. An increase in Ms results in geopolymer binders with better mechanical properties, but this is only true up to a certain value. This optimum Ms value varies for different geopolymer systems, based on the types of source materials used [16,75,81,82].

Increasing the activator content can also enhance the mechanical properties of a geopolymer by accelerating the geopolymerization reaction. However, similar to Ms, there does exist an optimum activator content, depending on the type of the precursor and activator [29,82]. Typical Ms values and activator contents related to common precursors, such as fly ash and slag, cannot simply be carried over to other waste precursors [75]. According to Sassoni et al. [58], a WCB geopolymer reaches a good geopolymerization level with a Ms of 1.4. Based on the optimum WCB geopolymer mixtures found in the literature (Table 2), this review identifies a representative Ms in the range of approximately 0.6 to 2.0 as being ideal for WCB-based geopolymers.

Fořt et al. [57] produced WCB geopolymer by combining powdered brick waste with a combined NaOH-Na$_2$SiO$_3$ activator and investigated the effect of activator concentration on its microstructure. They discovered that increasing the Ms decreased the compactness of the structure. At the same time, the specific pore volume increased at higher Ms ratios. The most compacted structure and highest reaction rate were seen in samples with Ms = 0.8. Cracks in the microstructure of the geopolymer samples were observed when the combined NaOH–Na$_2$SiO$_3$ Ms was higher, between 1.0 and 1.2 [57].

To evaluate the effect of alkali activator concentration on the mechanical properties of geopolymer mortar, Tuyan et al. [18] tested samples with different activator Ms ratios and different Na$_2$O contents. When the Na$_2$O content was increased from 4% to 6% (Ms 1.6 to 1.8) the compressive strength increased since the addition of Na$_2$O increased the alkalinity of the activator solution, allowing more Si$^{4+}$ and Al$^{3+}$ ions to dissolve [18,83]. Tuyan et al. [18] obtained a maximum compressive strength of 17 MPa and a minimum compressive strength of 3.8 MPa for the mixtures with 1.8 Ms and 10% Na$_2$O, and 1.6 Ms and 4%Na$_2$O, respectively. Further increasing the Ms beyond 1.8 resulted in a decrease in compressive strength.

Silva et al. [22] suggested an inverse relationship between the compressive strength of WCB geopolymers and Ms. According to Barbosa et al. [84], this is the result of the residual sodium silicate retarding the water evaporation in the mixture, thereby forming polymerization products during the polycondensation process. A 7-day compressive strength of 0.6 MPa was recorded for the geopolymer with Ms 1.50; the strength increased to 1.9 MPa when the Ms was reduced to 0.50 (in these samples, the Na$_2$O content, liquid/binder ratio, oven curing temperature, and oven curing time were fixed at 8%, 0.30, 65 °C, and 1 day, respectively) [22]. When optimizing the Ms value and Na$_2$O content, a maximum compressive strength of 21.3 MPa was recorded for the WCB geopolymer with Ms 0.6 and 8% Na$_2$O (the liquid/binder ratio was 0.29 and the curing time was 7 days at 65 °C).

Similarly, an optimum Na$_2$O content of 8% was suggested by Robayo-Salazar et al. [33]. The Na$_2$O concentration in geopolymer is important, in terms of balancing the electrical chargers in Al and Si tetrahedral structures and accelerating the geopolymerization reaction [84–86]. However, higher Na$_2$O contents can result in low compressive strengths because of the presence of unreacted alkalis, the formation of carbonates due to atmospheric carbonation, and efflorescence [29,32,87].

Instead of directly expressing the Na$_2$O content in the activator, some studies have given the NaOH content. Therefore, for NaOH also, there exists an optimum percent-
age above which the compressive strength significantly decreases [29,33,65]. Robayo-Salazar et al. [33] suggest 8% NaOH according to the weight of the binder as being the optimal content for WCB geopolymers. It follows that excessive NaOH in the mixture can cause depolymerization and the disassociation of the reaction products, while also weakening the geopolymer matrix via atmospheric carbonation [32,85]. Once the system attains the optimum hydroxide concentration, more Si and Al are added to the reaction medium, due to the higher dissolution rate, thereby forming aluminosilicate oligomers [12]. Subsequently, these products dehydrate and precipitate, forming aluminosilicate gels. At lower NaOH concentrations, retardation in the dissolution rate can be observed due to the insufficient alkalinity in the medium. In contrast, higher hydroxide concentrations dissolve previously formed aluminosilicate oligomers to form aluminate and silicate molecules, instead of allowing them to go on to form aluminosilicate gels [12,88].

In contrast, Ouda and Gharieb [35] claimed that there is a significant positive correlation between NaOH content and compressive strength. They argue that this is due to the fact that higher NaOH contents can offer more Na$^+$ ions to attack the aluminosilicates, thereby releasing more Si$^{4+}$ and Al$^{3+}$ ions for the geopolymerization reaction. They reported the highest compressive strength of 24.3 MPa at 28 days for the mixture with the highest NaOH content (12% of the total binder). A similar relationship was identified by Wang et al. [89] for a metakaolin-based geopolymer. However, none of the other literature on WCB-based geopolymers corroborates a similar relationship.

Migunthanna et al. [42] investigated the effect of solid activator (i.e., anhydrous Na$_2$SiO$_3$) content on the compressive strength development of WCB-based geopolymer binders. An activator content of 10% of the weight of the binder was identified as being optimum for the WCB-based binders. Lower activator contents are unable to provide sufficient alkalinity for the medium to facilitate the complete geopolymerization reaction, due to the co-precipitation of gel products without reaching the polycondensation phase [90]. On the other hand, higher alkaline contents facilitate the dissolution process, hence adding more silica to the medium. According to Escalante-García et al. [91], excess silica arising from the activator and the dissolution process can later form silicic acid, which can ultimately cause the deterioration of the geopolymerized structure.

The activator contents for WCB geopolymer syntheses related to Ms, Na$_2$O, and NaOH were reviewed in this section. In summarizing the current research findings, this review specifies an approximate range for Ms from 0.6 to 2.0 and Na$_2$O or NaOH as from 7% to 10% of the binder. In addition, considering the solid activators, 10% of Na$_2$SiO$_3$ from the weight of the total binder content is appropriate for the WCB-based geopolymers. Limited research, widely varying production conditions, and WCB sourcing variability make it difficult to establish a more precise range for the above parameters. Therefore, further studies on producing WCB-based geopolymer binders are essential to understanding the chemistry, optimum activator concentrations, and optimum degree of geopolymerization [30].

When producing geopolymer concrete for pavement construction, the use of both solid and aqueous activators is possible, depending on the application [68]. The use of a combined NaOH–Na$_2$SiO$_3$ aqueous activator, with Ms at 2 and Na$_2$O at 14.2%, was identified as suitable for pavement concretes produced using common ACMs, such as fly ash and slag [68]. However, these solutions may need to be optimized or modified for other aluminosilicate materials, particularly for materials such as WCB.

Generally, solid activators are more suitable for in-situ constructions (i.e., field applications at an ambient temperature) while aqueous activators are better for pre-cast applications [68]. Migunthanna et al. [43] experimentally showed the feasibility of using solid activators to produce WCB-based geopolymer concrete for pavement base and sub-base applications. Geopolymer concrete trial mixes, with a 28-day compressive strength in the range of 30 MPa to 40 MPa, were developed using WCB-based one-part binders with a Na$_2$SiO$_3$ activator. However, more studies on the development of geopolymer concrete and an assessment of their mechanical and durability performance are necessary to understand the possibility of using these low-carbon binders in the pavement industry.
6. Effect of Curing Conditions

Curing temperature and time directly affect the geopolymerization process and reaction rates, with elevated temperatures generally leading to better geopolymerization [22]. According to Robayo-Salazar et al. [33], a WCB geopolymer cured at 25 °C recorded a 28-day compressive strength of around 6.5 MPa, while specimens from the same mixture, cured at 70 °C for 24 hours and then at 25 °C for 27 days, showed an enhanced compressive strength of around 11.2 MPa. In this case, curing at an elevated temperature for only 24 h increased the compressive strength by 72%. However, long oven-curing times at high temperatures can decrease the density of geopolymer matrices due to higher water evaporation [22]. Therefore, it is necessary to carefully select the curing conditions to balance the effects of faster reaction with the effects of water evaporation [22,32].

Since WCB needs more time for geopolymerization than other aluminosilicate materials, such as fly ash and slag, WCB generally requires longer curing periods at elevated temperatures. Silva et al. [22] suggest curing for 7 days at 65 °C to 80 °C as the best curing conditions for WCB-based geopolymers. Similarly, Tuyan et al. [18] record an enhanced compressive strength by increasing the curing temperature from 50 °C to 80 °C and the curing period from 1 day to 7 days. Furthermore, the results by Komnitas et al. [32] and Ulugöl et al. [20] show that a temperature below 60 °C is not sufficient to produce a WCB geopolymer with noticeable strength.

The samples reported by Silva et al. [22], which were cured at 95 °C, showed no further strength development beyond 3 days of curing. Similarly, Tuyan et al. [18] showed there was no further strength development beyond 5 days for 90 °C and 100 °C curing temperatures. Further, Mahmoodi et al. [63] reported that the compressive strength of WCB geopolymer cured at 100 °C had a significant drop compared to samples cured at 75 °C. This may be attributed to the geopolymerization reaction stopping due to the rapid water loss from the matrix and reaction products deteriorating at high temperatures [18,22,92,93]. In fact, higher strengths can be achieved by longer curing periods at more moderate temperatures [22,32]. Through reaction heat evaluation, Fořt et al. [57] showed that geopolymer cured at 80 °C exhibited a second peak in its specific heat power vs time curve. They explain this second peak as corresponding to the formation of aluminosilicate gels and polymerization products. However, a similar peak was not identified at curing temperatures below 60 °C. From summarizing the experimental results available in the literature (shown in Table 2), temperatures around 80 °C can be suggested as the optimal curing temperature for WCB geopolymer.

A few studies have investigated the mechanical properties of WCB geopolymer cured at an ambient temperature (20 °C to 25 °C). Figure 6 shows the difference in the 7-day compressive strength of WCB geopolymer samples cured at elevated and ambient temperatures. It is clear that WCB geopolymers cured at elevated temperatures have higher compressive strength compared to those cured at ambient temperatures. Robayo-Salazar et al. [33,60] and Fořt et al. [57] recorded satisfactory 28-day compressive strengths of 41.9 MPa, 54 MPa, and 59 MPa, respectively, for geopolymer samples cured at ambient temperatures. However, these samples showed a very low early strength development. For example, according to Robayo-Salazar et al. [60], the 7-day strength of ambient-cured WCB geopolymer was 5 MPa, almost 88% less than its 28-day strength. Likewise, Tuyan et al. [18] recorded compressive strengths of 0, 0, 5.3 MPa, and 18.7 MPa at 3, 7, 28, and 90 days, respectively, for WCB geopolymers cured at ambient temperature (25 °C). Similarly, for one-part WCB geopolymers subjected to ambient curing, Migunthanna et al. [42] reported around 1 MPa, 3 MPa, 5.3 MPa, and 10.5 MPa of compressive strength in 3, 7, 28, and 90 days, respectively. These results imply that for 100% WCB-based geopolymers, it is more suitable to use elevated curing temperatures to achieve high compressive strength, especially for early strength. Therefore, the use of WCB as the sole precursor for developing pavement concretes is not practically feasible.
Figure 6. The 7-day compressive strength of WCB geopolymer, cured at elevated and ambient temperatures. In all cases, a combined NaOH–Na$_2$SiO$_3$ activator was used. Note: $t = $ curing temperature and amb. temp = ambient temperature [18,22,29,30,32,41,60].

Calcined aluminosilicate materials, such as fly ash, slag, metakaolin, and calcined natural pozzolans, such as clays, are identified as suitable ACMs for producing base and subbase concrete [68,94,95]. Since WCB is a calcined clay-based material, it is a possible alternative for use in pavement concrete as a cementitious binder or stabilizer. According to the current literature on WCB geopolymer binders, higher curing temperatures are essential to achieve satisfactory strength values. However, in field applications, the provision of elevated temperatures is difficult in practice and cost-ineffective. Hence, it is suggested not to use WCB as the sole aluminosilicate precursor for pavement concretes. Similarly, Austroads general guide for geopolymer concrete [68] also highlights that geopolymer concretes based on fly ash and metakaolin alone are not suitable for base and subbase applications. The guidelines further suggest blending these types of precursors with sources of calcium, such as slag and OPC, and then using them as a pavement construction material. Migunthanna et al. [42,43] experimentally evaluated the suitability of WCB-based binders to produce pavement concretes and proposed using blends of WCB with slag and fly ash to achieve the required strengths at ambient curing temperatures.

7. Binary and Ternary Geopolymer Systems with WCB and Other Aluminosilicate Materials

With the appropriate activator types, activator content, and curing conditions, a geopolymer with WCB alone showed 7- and 28-day average compressive strengths of around 20 MPa to 30 MPa (Table 2). However, the geopolymer showed very low or no strength development at ambient curing conditions. Since WCB is a relatively less reactive and less amorphous precursor, a WCB-alone geopolymer requires more time to develop its mechanical properties [22,41].

Binary geopolymers made from WCB blended with other aluminosilicate materials, such as fly ash [41,42,61,64], slag [30,42], metakaolin [59], and OPC [33,60], have a high potential for giving better mechanical properties than a geopolymer with WCB alone. For example, the 90-day compressive strength of a WCB geopolymer cured at ambient
temperatures was 15 MPa, which increased to 83 MPa after replacing 60% of the WCB with slag [30]. Furthermore, binary blends of WCB with slag, and ternary blends of WCB with fly ash and slag, have already been identified as feasible precursor combinations to develop pavement-grade concretes [43]. Figure 7 further illustrates the difference between the compressive strengths of ambient-cured WCB-alone geopolymers and those of blended geopolymers. When compared to WCB-alone geopolymers, these blends of WCB-based geopolymers generally show a 60–93% increase in compressive strength, depending on the mix proportion. The optimal compositions of the blended precursors, curing conditions, and activators recorded in the literature are summarized in Table 3.

Figure 7. Comparison of the 7-day compressive strengths of ambient temperature-cured 100% WCB-based geopolymers and binary and ternary geopolymers with other aluminosilicate materials. Note: FA = Fly ash [30,41–43,60].

Rovnanik et al. [41] investigated WCB- and fly ash-based geopolymers cured at room temperature and obtained a 7-day compressive strength of 21 MPa, for a sample with a WCB to fly ash ratio of 1:3. The same combination achieved a compressive strength of 40 MPa and 48 MPa at 28 and 90 days, respectively (Figure 8). Modifying the WCB and fly ash mix proportions to 1:1 did not make a significant change to the compressive strength, being around 20 MPa, 46 MPa, and 48 MPa at 7, 28, and 90 days, respectively. Similarly, Wong et al. [64] investigated replacing the fly ash with WCB at lower replacement quantities of 10% and 20% when producing geopolymer mortar. They reported that an optimum 10% replacement of fly ash with WCB achieved a strength of 44.2 MPa at 28 days. The 28-day compressive strengths reported in both studies showed no significant difference with 10% and 50% replacements of fly ash with WCB. A similar relationship was observed for one-part geopolymers synthesized with WCB and fly ash [42]. Increasing the fly ash content beyond 40% did not cause a significant improvement in compressive strength.
Table 3. Binary and ternary geopolymer binders with WCB and other aluminosilicate materials, as recorded in the literature: composition of the binary precursors, curing conditions, activators, and compressive strengths.

<table>
<thead>
<tr>
<th>Reference</th>
<th>Maximum Compressive Strength (MPa)</th>
<th>Days</th>
<th>Curing Temperature (°C)</th>
<th>Relative Humidity (%)</th>
<th>Activator</th>
<th>Activator Content (by % Weight of Binder)</th>
<th>Liquid /Binder Ratio</th>
<th>Composition of the Precursor</th>
</tr>
</thead>
<tbody>
<tr>
<td>Rovnanik et al. [41]</td>
<td>21 ± 1 20 ± 1.5</td>
<td>7</td>
<td>21 ± 2</td>
<td>50 ± 2</td>
<td>NaOH + Na₂SiO₃ (activator Ms = 1.0)</td>
<td>25%</td>
<td>0.36</td>
<td>75% fly ash + 25% WCB</td>
</tr>
<tr>
<td>Guo et al. [61]</td>
<td>Not assessed</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>70% fly ash + 30% WCB</td>
</tr>
<tr>
<td>Wong et al. [64]</td>
<td>44.2</td>
<td>28</td>
<td>amb.t</td>
<td>-</td>
<td>NaOH + Na₂SiO₃ (Wt. Na₂SiO₃/NaOH = 2.5)</td>
<td>10M NaOH</td>
<td>0.25</td>
<td>90% fly ash + 10% WCB</td>
</tr>
<tr>
<td>Migunthanna et al. [42]</td>
<td>~10 22.4</td>
<td>7</td>
<td>25 ± 2</td>
<td>-</td>
<td>Na₂SiO₃ (solid)</td>
<td>10%</td>
<td>0.32</td>
<td>60% fly ash + 40% WCB</td>
</tr>
<tr>
<td>Zawrah et al. [30]</td>
<td>~65 ~78</td>
<td>7</td>
<td>25 ± 2</td>
<td>-</td>
<td>NaOH + Na₂SiO₃ (Vol. Na₂SiO₃/NaOH = 2.5)</td>
<td>30%</td>
<td>0.30</td>
<td>60% slag + 40% WCB</td>
</tr>
<tr>
<td>Migunthanna et al. [42]</td>
<td>68.3 81.8</td>
<td>7</td>
<td>25 ± 2</td>
<td>-</td>
<td>Na₂SiO₃ (solid)</td>
<td>10%</td>
<td>0.32</td>
<td>60% slag + 40% WCB</td>
</tr>
<tr>
<td>Rovnanik et al. [59]</td>
<td>23 ± 4</td>
<td>7</td>
<td>40 °C for 20 h, after, at amb. temp</td>
<td>-</td>
<td>NaOH + Na₂SiO₃ (act Ms = 1.4)</td>
<td>-</td>
<td>-</td>
<td>75% metakaolin + 25% WCB</td>
</tr>
<tr>
<td>Ouda and Gharieb [35]</td>
<td>~39 ~34 ~50</td>
<td>7</td>
<td>80 °C for 24 h, after, at 40 °C</td>
<td>&gt;95</td>
<td>NaOH</td>
<td>12 M</td>
<td>0.243</td>
<td>20% waste concrete powder + 80% WCB</td>
</tr>
<tr>
<td>Robayo-Salazar et al. [33]</td>
<td>41.39 102.59</td>
<td>28</td>
<td>25</td>
<td>90</td>
<td>NaOH + Na₂SiO₃ (act Ms = 1.3)</td>
<td>Na₂O = 8%</td>
<td>0.23</td>
<td>10% OPC + 90% WCB</td>
</tr>
<tr>
<td>Robayo-Salazar et al. [60]</td>
<td>26 ± 2 34 ± 0.5 14 ± 1 22 ± 2 22 ± 1 42 ± 5 28 ± 2 35 ± 1 57 ± 3 102 ± 14</td>
<td>7</td>
<td>25 ± 3</td>
<td>80</td>
<td>Na₂SO₄</td>
<td>6%</td>
<td>0.23</td>
<td>30% OPC + 70% WCB</td>
</tr>
<tr>
<td>Migunthanna et al. [43]</td>
<td>~54 ~75 ~65 ~89 ~66 ~92</td>
<td>7</td>
<td>25 ± 2</td>
<td>-</td>
<td>Na₂SiO₃ (solid)</td>
<td>10%</td>
<td>0.32</td>
<td>10% fly ash + 36% slag + 54% WCB</td>
</tr>
</tbody>
</table>


As shown in Figure 9a, the rate of compressive strength development reported for the binary blends of WCB and fly ash with 1:3 and 1:1 replacement ratios showed a similar trend. The same behavior was also seen in the experimentally measured flexural strength values (Figure 9b). The 7-day flexural strength was similar in 75% and 50% replacements of WCB with fly ash (approximately 7 MPa) and was around 30% higher than fly ash alone.

Generally, fly ash as a sole binder reacts very slowly at ambient conditions and does not achieve high strengths [96]. Therefore, blindly increasing the fly ash content will not
keep improving the strength of the blended mixture. However, all the fly-ash-blended mixtures investigated by Rovnaník et al. [41] and Migunthanna et al. [42] showed better strengths than the pure WCB geopolymer. This can be attributed to the higher pozzolanic activity and higher specific surface area of fly ash compared to WCB. Furthermore, the addition of fly ash to WCB reduces the shrinkage of the mixtures since fly ash absorbs less water compared to WCB [41].

Zawrah et al. [30] studied the feasibility of using WCB and slag to synthesize geopolymers at ambient curing conditions. The addition of slag increased the bulk density and decreased the porosity and water absorption of the geopolymer samples, indicating better geopolymerization. For example, in ambient curing conditions, the 3-day compressive strength was improved from around 5 MPa to 30 MPa by replacing 40% of the WCB with slag [30]. They obtained the highest compressive strengths with a 60% replacement of WCB with slag; the strengths were around 53 MPa, 65 MPa, 78 MPa, and 83 MPa at 3, 7, 28, and 90 days, respectively (Figure 10a). Compared to the 100%WCB geopolymer, the blend of WCB with slag showed a higher rate of strength development at all ages (Figure 10b). Similar behavior was observed in a one-part geopolymer with WCB and slag. According to Migunthanna et al. [42], a 60% replacement of WCB with slag was identified as the optimum, recording compressive strengths of around 43 MPa, 68 MPa, 82 MPa, and 89 MPa at 3, 7, 28, and 90 days, respectively.

Figure 10. (a) Compressive strength development of 100% WCB geopolymer and a binary blend of WCB and slag. (b) Comparison of the rate of strength development in 100% WCB geopolymer and binary blends of WCB with fly ash and slag. (Source of data: Graphs A and C—Zawrah et al. [30]; Graph B—Rovnaník et al. [41]).

In contrast to WCB–fly ash blends, a significantly higher initial strength development rate was identified in the blends of WCB with slag. However, after 7 days, both blends showed a similar trend in strength gain (Figure 10b). The high CaO content in slag results in the formation of calcium silicate hydrates alongside the aluminosilicate matrix, which reinforces the geopolymer gel and improves its strength [30,97,98]. Moreover, the calcium species present in the precursor provide nucleation sites for dissolved materials and reduce the total setting time [35,99]. However, higher replacement percentages of WCB with slag intensified the formation of micro-cracks and the samples deteriorated [30,42]. This can be attributed to the shrinkage of geopolymer gels during the hardening process [35].
In addition to blending WCB with other precursors, Migunthanna et al. [42,43] demonstrated that geopolymers with better mechanical, microstructural, and mineralogical properties can be developed by using the one-part mixing method, eliminating the NaOH from the activator, and using solid Na$_2$SiO$_3$ as the sole activator. Figure 11 shows the comparisons of compressive strength and the rate of strength development for these geopolymers. The case of the blends of WCB with fly ash, the removal of NaOH from the activator reduced the compressive strength. Fly ash is a very low reactive precursor under ambient temperatures; therefore, Na$_2$SiO$_3$ cannot provide the necessary alkalinity to facilitate the dissolution of silica from fly ash [100]. However, a significant effect on the compressive strength was not observed for the WCB–slag blends after removing the NaOH from the activator and using solid Na$_2$SiO$_3$ as the sole activator.

**Figure 11.** Comparisons of (a) compressive strength and (b) the rate of strength development for one-part and two-part geopolymers, synthesized from WCB blended with fly ash or slag (source of data: Zawrah et al. [30], Rovnanik et al. [41] and Migunthanna et al. [42]).

WCB–metakaolin mixtures can also be used as an aluminosilicate precursor to produce geopolymers [59]. The compressive strength of geopolymer increased from 10.2 MPa to 23 MPa after replacing 75% of the WCB with metakaolin [59]. This strength increment is approximately 17% higher than the 7-day compressive strength of geopolymer with metakaolin alone. The flexural strength of geopolymers with blends of WCB and metakaolin increased with the increase in the metakaolin content [41,59]. Replacing 75% of the WCB with metakaolin resulted in a 7-day flexural strength increment from 0.78 MPa to 4.5 MPa. Compared to blending WCB with slag, WCB–metakaolin blends showed lower compressive strength values. This can be attributed to the slow early strength development of metakaolin in ambient conditions [101]. After 28 days, metakaolin as a binder does not show a significant difference in compressive strength between ambient and elevated curing conditions [101].

Ouda and Gharieb [35] analyzed the mechanical and microstructural properties of geopolymers prepared by substituting WCB with waste concrete powder and calcined waste concrete powder by up to 30%. All these specimens were cured at 80 °C for 24 h and then at 40 °C until the test date. They used NaOH as the alkali activator. The highest compressive strength at 7 days was recorded to be 39 MPa, with a 20% replacement with concrete powder. For all curing times, a 30% replacement with concrete powder generally...
resulted in lower strength values than a 20% replacement. However, when WCB was replaced by calcined concrete powder, the highest compressive strengths were noticeable in mixtures having a 5% replacement, with a maximum compressive strength of around 50 MPa at 28 days. Compressive strengths of around 25 MPa, 34 MPa, 31 MPa, and 25 MPa were recorded at 7 days for 5%, 10%, 15%, and 20% replacements with calcined concrete powder, respectively. After 14 and 28 days, the 10%, 15%, and 20% mixtures showed lower strength values than the 50 MPa achieved at a 5% replacement. This can be attributed to the reduction in the soluble alumina and silica in the mixtures with more than 5% of the calcined concrete powder. However, by evaluating the strength increment, Ouda and Charieb [33] suggest that concrete waste most likely acts as micro-aggregates rather than contributing to the geopolymer matrix [35].

Robayo-Salazar et al. [33] produced a WCB geopolymer with the addition of small amounts of OPC. This addition significantly increased the compressive strength of the samples, while also removing the necessity of thermal curing. These effects can be explained as the CaO, present in OPC, producing cement hydration products that coexist alongside the aluminosilicate gels [96,102]. A maximum 28-day compressive strength of 102.59 MPa was recorded for WCB with a 20% OPC replacement and a combined NaOH-Na$_2$SiO$_3$ activator [33]. For a geopolymer activated with only NaOH, a maximum compressive strength of 41.39 MPa was achieved [33].

The compressive strength of the combined WCB–OPC precursors, activated with Na$_2$SO$_4$ and Na$_2$CO$_3$, increased with the increase in OPC content in the precursor. This can be attributed to the formation of more OPC hydration products than aluminosilicate gels [103]. However, the strengths of the precursors activated with Na$_2$SO$_4$ and Na$_2$CO$_3$ generally showed lower maximum compressive strengths than the specimens activated with NaOH and combined NaOH–Na$_2$SiO$_3$ [33]. The Na$_2$SiO$_3$ employed in the activator has the ability to donate soluble silicas to the geopolymerization process [78], while such an ability is not present in the activators that contain Na$_2$SO$_4$ and Na$_2$CO$_3$.

Further, Migunthanna et al. [43] developed ternary geopolymer systems by blending WCB with both slag and fly ash. In these binders, the maximum replacement of WCB with fly ash was limited to 30%. All blends were capable of achieving more than 50% of their 28-day compressive strength within the first 72 h. The addition of fly ash to the WCB and slag mixture facilitated a workable and homogenous mixture, while also reducing the shrinkage [41,104].

Although high-temperature curing is more suitable for 100% WCB-based geopolymers, higher compressive strength can be achieved even at ambient temperatures by blending WCB with other aluminosilicate materials. According to the experimental values available in the literature (Table 3), it is clear that binary and ternary geopolymer systems are superior to a 100% WCB-based geopolymer in many aspects.

The experimental results of binary geopolymer systems of WCB with slag or OPC and ternary blends of WCB, slag, and fly ash show high compressive strength and high early strength development at ambient curing, compared to other blended aluminosilicate precursors. Therefore, WCB–slag, WCB–OPC, and WCB–slag–fly-ash blends can be proposed as suitable binding materials for use in the pavement base and subbase concrete. Mixing low-calcium precursors with high calcium sources, such as lime, cement, slag, and calcium aluminate is already recommended in the guidelines [68].

Although there is research available on the development of WCB-based binders, experimental data on the development of concrete using low-carbon binders are very limited. To the best of the knowledge of the authors, only Migunthanna et al. [43] have studied the development of concrete with WCB-based binders. The study shows the feasibility of producing a geopolymer concrete with WCB-based binary and ternary binders to use in pavement base and subbase concrete. They investigated twelve concrete mix designs using four binder types and three aggregate combinations, as shown in Table 4. Concrete with a 28-day compressive strength of around 38 MPa was developed using a WCB–slag binder (i.e., Binder 1). By using WCB–slag–fly-ash binders, compressive strengths of up
to 41 MPa were achieved. However, the mechanical performance and durability of these materials should be comprehensively assessed prior to any field applications.

8. Effect of the Liquid-to-Binder Ratio

Similar to OPC binders, the strength of geopolymer binders also has a strong relationship with its liquid-to-binder (l/b) ratio. The optimum l/b ratio depends on the type of aluminosilicate material used [22,29]. For example, the optimum l/b ratio of WCB geopolymer is decreased when partially replacing raw WCB in the precursor with fly ash [41,42]. The lower liquid content in the mixture decreases the pore volumes and shrinkage cracks, resulting in enhanced mechanical properties [105]. According to Tuyan et al. [18], decreasing the l/b ratio from 0.46 to 0.40 improved the 7-day compressive strength of WCB geopolymer mortar by 24%. Similarly, by decreasing the l/b ratio from 0.45 to 0.30, Reig et al. [29] increased the compressive strength from 30 MPa to 50 MPa (Table 3). However, the excessive lowering of liquid content can reduce the strength, due to the presence of unreacted particles and the resulting poor cohesiveness [22,42].

Higher l/b ratios naturally mean that the water content in the geopolymer mixture is also higher. Excess water in the paste that is not consumed in the hydration process increases the porosity of the hardened geopolymer, leading to weak compressive strength [106]. Specifically, when the activator contains NaOH, increasing the NaOH content can reduce the water required for driving the geopolymerization reaction, without significantly affecting the workability [35]. For example, increasing the NaOH content by 6% reduces the required water content by 9%, while maintaining its workability [35]. However, when increasing the NaOH content, it is important not to exceed the optimum NaOH dosage, as discussed in the previous section.

Good workability of the fresh concrete mixture allows easier placement and proper finishing of the pavement during its construction [9]. Similar to the binders, the required workability of WCB geopolymer concrete can also be achieved by blending low water-absorbing source materials with WCB and using proper activators. Migunthanna et al. [43] show that when developing pavement concrete with WCB binders, the addition of fly ash can reduce the water-to-binder ratio required to achieve the target slump value.

In addition, superplasticizers can further assist in increasing workability. However, some superplasticizers are intolerant of highly alkaline mediums, while some are weak due to clay minerals [68,107]. Naphthalene-based sulfonated superplasticizers have been identified as being suitable for use in highly alkaline mediums [68]. Superplasticizers belonging to lignosulfonates and polymelamine sulfonate can also be used with clay minerals [107]. However, further laboratory studies are required to identify the suitability and practicality of the various superplasticizers for different WCB blends.

9. Mineralogical and Microstructural Properties of WCB Geopolymers

Similar to raw WCB, quartz was the major mineral phase present in WCB geopolymers [18,28,29,32,42,57]. Clear peaks, corresponding to albite and muscovite, were also present in the samples [18,28,29,32,42,57]. These phases indicate partial geopolymerization and the presence of unreacted silica in the end products. However, the intensity of the quartz and muscovite crystal peaks were slightly diminished, compared to raw WCB [18,30,32,57]. New peaks corresponding to the different hydration products were
slightly visible [30]. These peaks were consistently recorded in mixes with high compressive strengths and in mixes cured at high temperatures, indicating the formation of geopolymerization products [18,30,42].

Komnitsas et al. [32] recorded a pirssonite (Na₂Ca(CO₃)₂·2H₂O) phase in their WCB geopolymer samples. According to their findings, this phase was generated as a result of atmospheric carbonation. Foť et al. [57] observed the formation of zeolitic phases for geopolymers cured at 60 °C and 80 °C. Geopolymer binders with WCB and slag showed semi-crystalline phases, such as the calcium silicate hydrate phase and sodium calcium aluminosilicate hydrate phase. However, the features of the XRD pattern of the WCB geopolymer samples were similar to that of raw WCB [30,43].

The high content of alumina and silica in bricks helps to develop noticeable degrees of homogeneity in the WCB-based geopolymer [32]. The scanning electron microscopic images obtained by Zawrah et al. [30] showed the formation of geopolymer products, indicating the capability of WCB as a source material to synthesize geopolymer. However, the angular shape of WCB particles (Figure 12a) makes the microstructure less compacted and more porous [41,43]. Figure 12b,c further verifies that, irrespective of the curing temperature, a WCB-only geopolymer has a porous and disturbed microstructure, with an abundant presence of unreacted particles. This less compact microstructure suggests that WCB alone cannot serve as source material to synthesize high-performance geopolymer [35]. Less porous and denser products can be obtained by mixing WCB with other precursors, such as fly ash, slag, or OPC (Figure 12d) [30,33,42,43,61].

All these results justify, from a microstructural viewpoint, the argument that WCB, blended with other aluminosilicate materials, should indeed improve the mechanical properties and the durability of geopolymers.

10. Structural Integrity and Durability of WCB-Based Geopolymers

Vafaei and Allahverdi [62] investigated the acid resistance of geopolymer mortar, using WCB and phosphorous slag in the binder. The samples were immersed in sulfuric acid (H₂SO₄) and hydrochloric acid (HCl), with a pH of 1, for six months. The study by Vafaei et al. [108] further investigated the change in compressive strength, mass, and appearance of a geopolymer mortar produced with similar precursors and exposed to
mild acid concentrations (pH = 3) for a 24-month period. Both these studies showed that compared to OPC mortar, WCB-based mortar has an excellent durability performance with respect to acid attack. For instance, the mass loss observed in WCB-based mortar exposed to HCl (pH = 3) for 24 months was 4.5%, while OPC mortar showed a 16% loss under the same conditions. In the same experiment, the compressive strengths deteriorated by 2.6% and 68% for WCB-based and OPC-based mortars, respectively [108]. Table 5 further summarizes the strength and mass reduction of WCB- and OPC-based mortars exposed to an acid environment, as reported in these two studies.

Table 5. Compressive strength and mass loss of WCB mortar and OPC mortar exposed to different acids.

<table>
<thead>
<tr>
<th>Reference</th>
<th>Acid</th>
<th>Mass Loss</th>
<th>Compressive Strength Loss</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>WCB Mortar</td>
<td>OPC Mortar</td>
</tr>
<tr>
<td>Vafei et al. [108]</td>
<td>HCl, pH = 3</td>
<td>4.5% after 720 days</td>
<td>16% after 720 days</td>
</tr>
<tr>
<td></td>
<td>H₂SO₄, pH = 3</td>
<td>43.4% after 720 days</td>
<td>56.6% after 720 days</td>
</tr>
<tr>
<td>Vafaei and Allahverdi [62]</td>
<td>HCl, pH = 1</td>
<td>36% after 180 days</td>
<td>48% after 180 days</td>
</tr>
<tr>
<td></td>
<td>H₂SO₄, pH = 1</td>
<td>34% after 180 days</td>
<td>58% after 180 days</td>
</tr>
</tbody>
</table>

Komnitsas et al. [32] showed that the WCB geopolymer has satisfactory resistance to freeze-thawing by subjecting them to freeze-thaw cycles from −10 °C to 60 °C. In this study, geopolymers were subjected to weekly freeze-thaw cycles for two months and recorded a compressive strength reduction from 49.5 MPa to 48 MPa. However, when the WCB geopolymer samples were immersed in water for two months, the compressive strength was reduced by 55% [32]. According to Komnitsas et al. [32], this loss of strength resulted from the depolymerization of the aluminosilicate network by higher water absorption during the immersion period.

According to Komnitsas et al. [32], the WCB geopolymer strength reduced from 49.5 MPa to a minimum value of 37 MPa after heating at 800 °C. Contrasting these results, Tuyan et al. [18] showed that WCB geopolymer had a thermally stable structure upon heat treatment of up to 1000 °C. Considering the positive heat resistance ability of WCB geopolymer, Tuyan et al. [18] suggest using WCB geopolymer as an alternative to OPC in manufacturing elements that may be exposed to high temperatures.

Generally, WCB geopolymers do not show significant weight loss under heat treatment [18]. According to Tuyan et al. [18], a notable weight loss only occurred at temperatures of between 100 °C and 200 °C, corresponding to the evaporation of water from the matrix. Similarly, several other studies reported a peak weight loss at around 95 °C to 200 °C, corresponding to the evaporation of free and loosely bound water within the geopolymer matrix [29,57,109,110].

The curing conditions and the activator content incorporated during the geopolymer production also affect weight loss under heat treatment. Significant weight loss was found in the mixtures with higher NaOH dosage [57]. Unreacted NaOH forms sodium carbonates by atmospheric carbonation, which subsequently decompose when exposed to high temperatures [18,29]. Consequently, the geopolymer indicates a weight loss of around 400 °C, corresponding to the decomposition of these carbonates [111]. Tuyan et al. [18] recorded the weight loss corresponding to the decomposition of carbonates to be 650–750 °C. WCB geopolymer samples that were activated incorporating Na₂SiO₃ showed a low mass loss between 200 °C and 800 °C, indicating better geopolymerization [29].

Moreover, the porosity of the WCB geopolymer reduced with the addition of Na₂SiO₃ to the mixture [29]. Increasing Ms from 0 to 1.6 decreased the porosity from 2.02% to 1.83%. Further increasing the Ms up to 2.2 increased the porosity to 1.94% [18]. Samples activated with NaOH showed a higher porosity when compared to samples activated with the combined NaOH–Na₂SiO₃ [29]. Low porosity is an indication of high compressive strength and durability since it reduces the penetration of aggressive chemicals into the microstructure [42,61]. Therefore, in terms of durability, Reig et al. [29] recommend using Na₂SiO₃.
in the activation of WCB. However, more research investigating durability is essential to understand the possible applications and performance of WCB-based geopolymers.

The durability of pavement concrete is generally assessed based on tests corresponding to shrinkage, alkali-aggregate reaction, carbonation, chloride penetration, sulfate attack, freeze-thaw, and abrasion [80,112,113]. Although these test methods are well-developed and are specified for OPC concrete, they cannot be applied directly to evaluate the durability of geopolymer concrete [68,114]. Therefore, new test methods to assess the durability are needed, not only for WCB-based binders but also for geopolymer concrete produced with any type of precursors. However, Provis and Van Deventer [114] suggest that several existing test methods for conventional concrete appear suitable for geopolymers, with slight modifications where necessary, for example, by changing only the sample preparation and curing stages without modifying the test method itself.

11. Summary and Conclusions

The development of a WCB-based geopolymer is an innovative step in producing low-carbon binders, while also providing a profitable solution for solid waste management. WCB is a calcined clay-based material and has high potential to serve as an aluminosilicate precursor. WCB can be recovered from C&D waste and from waste generated during the brick manufacturing process.

Quartz is present as the dominant mineralogical phase in raw WCB and generally shows a low amorphousness compared to other commonly used aluminosilicate source materials, such as fly ash and metakaolin. Of the amorphous compounds present in raw WCB, SiO$_2$ and Al$_2$O$_3$ form the majority. The particle size of raw WCB significantly affects the mechanical properties of the resulting WCB-based geopolymer. Finer particles more easily react during geopolymerization and, therefore, result in higher compressive strength.

NaOH and combined NaOH–Na$_2$SiO$_3$ are the most common activators used in WCB-based geopolymer synthesis. Only a few studies are available on using Na$_2$CO$_3$–Na$_2$SO$_4$ and Na$_2$SiO$_3$-only activators. The soluble silica present in Na$_2$SiO$_3$ encourages the geopolymerization process and enhances the geopolymer’s mechanical properties. Therefore, most studies recommend incorporating Na$_2$SiO$_3$ in the activation of WCB. The use of NaOH in activators can positively affect the geopolymerization process by allowing the dissolution of more Si$^{4+}$ and Al$^{3+}$ ions. However, there is an optimum NaOH concentration, above which the compressive strength decreases because of the presence of unreacted alkalis, carbonate formation by atmospheric carbonation, and efflorescence.

Curing conditions strongly affect the geopolymerization of WCB, and elevated curing temperatures are generally preferred. WCB-based geopolymers cured at ambient temperatures showed very low or no strength development. All the published studies agree that temperatures above 60 °C result in higher compressive and flexural strengths compared to the samples cured at lower temperatures.

Binary and ternary geopolymer systems that compound WCB with other aluminosilicate materials, such as slag, fly ash, metakaolin, waste concrete powder, and OPC showed better performance than the 100% WCB-based geopolymer. The mechanical properties of these hybrids are generally superior in terms of compressive strength and thermal curing is not so necessary for strength development. From these blends, WCB–OPC, WCB–slag, and WCB-slag–fly-ash samples exhibit the highest compressive strengths since the CaO present in both OPC and slag encourages the formation of calcium silicate hydrates, along with the aluminosilicate matrix, thereby reinforcing the geopolymer gel.

WCB geopolymers possess a thermally stable structure under heat treatment and the main weight losses are attributed to the evaporation of free or loosely bound water and to the decomposition of carbonates. Blending with other aluminosilicate materials and the use of Na$_2$SiO$_3$ in the activation reduced the porosity, indicating better mechanical properties and durability.

According to the Australian pavement design guidelines, concrete intended for use in rigid pavement base construction should have a minimum 28-day characteristic com-
pressive strength of 32 MPa. In subbase applications, lean-mix concrete requires the 28-day compressive strength to be not less than 5 MPa. The study by Migunthanna et al. [43] showed that it is possible to develop concrete meeting the above strength requirements under ambient curing by using the WCB-based binders. In addition, WCB-based geopolymer binders showed low porosity and satisfactory resistance to freeze-thawing, which are critical considerations in rigid pavements.

By analyzing the state-of-the-art in this area, the following conclusions were drawn:

- In terms of the chemical and mineralogical composition, WCB has the potential to serve as an aluminosilicate precursor to synthesize geopolymer binders.
- An optimal Ms in the range of 0.6–2.0 and a Na₂O content of 7%–10% of the binder weight are ideal for a WCB geopolymer.
- The incorporation of Na₂SiO₃ in the activation is favorable in terms of improving both the strength and durability, compared to NaOH alone, Na₂SO₄, and Na₂CO₃.
- A curing temperature of 80 °C is identified as being optimal for producing the 100% WCB geopolymer.
- With ambient curing, WCB alone cannot achieve satisfactory strengths. Strength can be notably improved by using binary and ternary blends of WCB with other precursors, such as fly ash, metakaolin, slag, and OPC.
- Considering the high early strength gain, better long-term strength development, and a low-porous and compacted microstructure, the use of slag or OPC with WCB is recommended for developing pavement concretes. Incorporating fly ash is beneficial in situations where reducing shrinkage is critical.

Despite the research work to date, a comprehensive understanding of the role of WCB geopolymers in rigid pavements is still quite far away. For the successful implementation of these low-carbon binders, the following main research gaps need to be addressed in future studies:

- Conducting more comprehensive research on replacing hazardous activators, such as NaOH, and developing WCB binders using Na₂SiO₃ as the sole activator.
- More investigations on producing WCB-based one-part geopolymers, to find a more OPC-like alternative for the construction industry.
- Developing WCB-based pavement-grade geopolymer concrete (GPC) and optimizing the mix designs for use in pavement base and subbase constructions.
- Investigating the WCB binder and WCB-based GPC properties, such as strength (i.e., compressive and flexural) and durability (i.e., shrinkage, creep behavior, alkali–aggregate reaction, sulfate attack, permeability, and abrasion resistance).
- Conducting field studies with WCB-based GPC.

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