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| 1  | Future of Clay-Based Construction Materials- A review  |
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# 24 Abstract

Sustainability in the manufacture of different construction materials raises many important issues. Nowadays, there is increasing demand for such materials to be produced using environmentally friendly, low energy consuming production methods. This paper presents a review of the current research relating to the use of various production techniques for clay-based construction materials. The techniques which will be reviewed are: blending and stabilising, alkali activation (geopolymerisation) and the use of microwave heating as an innovative sintering, curing and drying method. The advantages and disadvantages of each technique will be discussed. Additionally, a comparison between the environmental and economic aspects of the studied production techniques along with some suggestions to improve the sustainability of different production techniques will be discussed. 

Keywords: Alkali activation; blending and stabilising; clay-based construction materials;
compressive strength; environmental impact; Geopolymerisation; microwave heating.

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# 46 **1. Introduction**

For many thousands of years, clay has been widely used as an integral part of construction
materials and products. Examples of the main structural clay products are bricks, blocks and
roof tiles. Floor and wall tiles are examples of non-structural products made from clay.
Buildings made from clay materials date back to the earliest periods of civilized development
[1, 2].

52 The desirable properties of clay-based products such as the durability, strength, heat and sound 53 insulation along with fire-resistance mean that there is still considerable demand for them in a 54 variety of sectors, despite the availability of modern alternative materials such as concrete, 55 glass-fibre/resin composites, steel and plastics [3].

Sun-baked clay bricks are thought to have been first used circa 8000 B.C while fired clay bricks where used circa 4500 B.C [4, 5]. In Europe, the Romans introduced clay-based brick during the 9<sup>th</sup> and 10<sup>th</sup> centuries and thousands of churches and cathedrals were built in masonry during the middle Ages [4]. The oldest skyscraper buildings in the world are located in the city of Shibam in Yemen [6]. These 500 huddled buildings, ranging from 5 to 11 stories high, reaching about 30m, were built with clay blocks [6, 7].

In 2013, the annual production of bricks was about 1391 billion units worldwide [5]. This 62 number is expected to increase through the rapid development of the construction industry 63 64 globally, together with an expected increase in the world population [8]. The conventional process of converting clay into brick involves firing the brick at temperatures ranging between 65 900 and 1150°C, depending on the type of clay [9, 10]. During this process, clay minerals break 66 down and sinter forming a glassy bond with other minerals and materials in the brick. The main 67 purpose of the firing process is to transform the porous and weak dried clay into strong, dense 68 69 bricks with low porosity [3, 11]. This process requires high levels of energy consumption and a resulting release of greenhouse gases into the atmosphere. The production of one brick
requires some 2.0 kWh of energy and the release of approximately 0.4 kg of CO<sub>2</sub> [9]. These
undesirable features of the manufacturing process are the main driving force behind research
into more sustainable alternatives [12].

New methods have been developed to produce alternative clay-based construction products with better performance and properties than those created in the firing process. One of the oldest techniques is the blending and stabilisation of clay with other cementing materials such as cement, lime and/or other waste materials [9, 13, 14]. Blending and stabilising clays with other waste or by-product materials has many benefits such as reducing land-fill, solving the issues of waste management, protecting the environment and saving energy, which in turn, reduces the cost of the final product [9].

81 In addition to the use of blended and stabilised clay-based construction products, researchers have investigated the use of alkali activation techniques (geopolymerisation). The concept of 82 83 developing clay-based geopolymer construction products is an attractive one, as they can provide structural strength in a very short time, they are sufficiently durable and CO<sub>2</sub> emissions 84 are reduced [15, 16]. Generally, geopolymer is formed by mixing an alumina-silicate precursor 85 with an alkali solution [17-21]. This technique relies on the chemical reaction between the 86 alumina-silicate precursor and a high alkaline solution to produce amorphous to semi-87 crystalline geopolymer [17, 21-23]. 88

Heat is essential for the curing, sintering and drying of clay-based construction products in order to gain an adequate strength for civil engineering applications [24-26]. However, the use of conventional heating methods is relatively slow due to the low thermal conductivity of claybased construction materials and the slow rate of heat transfer from their surface to their core [26, 27]. In addition, researchers have identified the disadvantages associated with 94 conventional heating technologies such as high-energy consumption, long processing times,
95 high processing temperatures and the associated negative environmental impacts [24, 26, 28,
96 29].

In the search for alternatives, innovative research has been carried out employing microwave 97 technology as a sintering, curing and drying technique in the production of clay-based 98 99 construction products [24-27]. The utilisation of microwaves has many advantages over conventional heating methods. Microwave treatment provides efficient internal heating, as 100 energy is supplied directly and penetrates the material through molecular interaction with the 101 electromagnetic field therein minimising substantial temperature gradients between the interior 102 and the surface [24, 30-34]. Microwave treatment reduces energy consumption via rapid 103 heating rates and processing times are significantly reduced leading to fewer negative 104 105 environmental effects. Physical and mechanical properties are also improved through resulting higher density materials with better grain distribution [24, 30-33]. 106

107 This paper presents a review of the research on the various techniques for the manufacture of clay-based construction products. The studied techniques including blending and stabilising, 108 alkali activation (geopolymerisation) along with the use of microwave heating are presented as 109 110 an innovative sintering, curing and drying technique in the production of clay-based construction products. Additionally, this paper also provide a comprehensive comparisons 111 regarding the environmental and financial aspects associated with different production 112 techniques along with some suggestions for future trend to improve the sustainability of the 113 studied production techniques. 114

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# 118 2. Review of Research

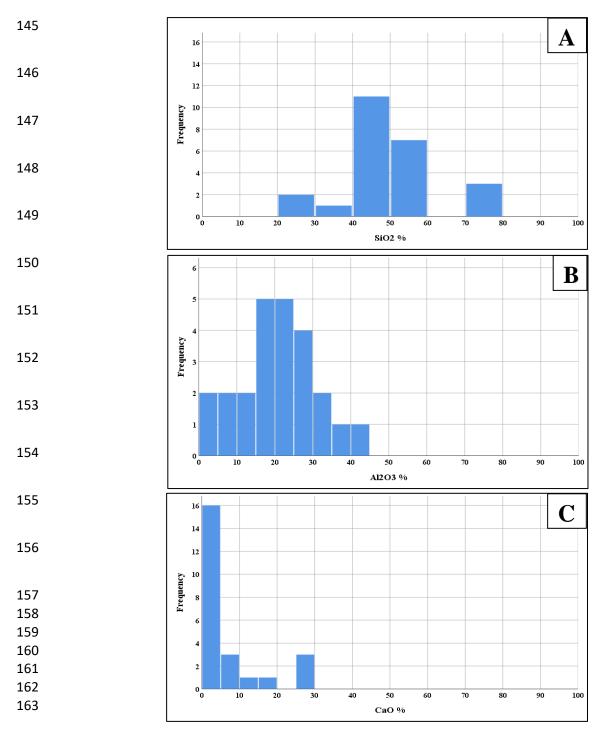
In this study, the production of different clay-based construction materials are divided into
three groups: blending and stabilising, alkali activation and microwave sintering, curing and
drying.

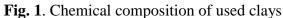
# 122 2.1 Characterization of the used clays

It is well understood that the chemical compositions of clay as raw material have significant 123 influence upon the properties of the clay-based construction materials [9]. Therefore, the 124 125 chemical composition of different types of clay should be tested to elucidate the performance of these materials when they are combined with stabilisers or being activated chemically by 126 different activators. The chemical composition of different types of clay as collected from the 127 128 Energy Dispersive X-ray Florescence Spectrometer (EDXRF) test for all the reviewed papers that providing such information are presented in Fig. 1. Fig. 1 displaying the ranges and 129 frequencies of the most common chemical compounds (SiO<sub>2</sub>, CaO and Al<sub>2</sub>O<sub>3</sub>) of the clay 130 powder materials used for the preparation of different clay-based construction materials. 131

It can be seen from Fig.1A that about 75% of the clays used in the production of different clay-132 133 based construction materials have SiO<sub>2</sub> content in the range between 40% to 60%. Additionally, 12.5% of the clays have showed  $SiO_2$  content in the range of 70-80 % and the other 12.5% 134 have SiO2 content in the range of 20-30%. The second chemical compound that can be found 135 136 in abundant a quantity in different types of clay is the Al<sub>2</sub>O<sub>3</sub>. Fig.1B shows that the data of the Al<sub>2</sub>O<sub>3</sub> content collected from different types of clay ranges from 0% to 45% and the majority 137 of the observations lies between 15% and 30%. Another important compound is the CaO 138 content. Fig.1C indicated that 67% of the clays have relatively small CaO content (below 5%), 139 while 25% of the clays have a CaO content in the range of 5-10% and 25-30% (12.5% for each 140 141 range).

As observed from Fig. 1 that most of the clays utilised in the production of different clay-based
construction materials displayed similar ranges of SiO<sub>2</sub>, CaO and Al<sub>2</sub>O<sub>3</sub> although they are came
from different origins and places around the world.





#### 166 2.2 Production of clay-based construction materials through blending and stabilizing

167 Stabilization is a process of mixing the clay with different types of binders with the aim of 168 enhancing its strength, durability and volume stability [35]. The performance of different 169 stabilised clay-based construction materials is depend upon the characteristics of soil, binder 170 and the mix design. This technique relies mainly on the formation of hydration products such 171 as C–S–H gel and C–A–S–H gel produced from the chemical reaction between the silica 172 sources (mainly the clay) and the CaO from the stabilisers.

In most of the reviewed papers, the clays have high percentages of SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>, while 173 having only small CaO content. Therefore, cement, lime and other binders with high CaO 174 content are added in small dosages to form C-S-H gel and C-A-S-H gel that enhancing the 175 176 mechanical and durability performance of the final product. The development of unfired clay-177 based construction products with comparable or better performance than fired products significantly reduces CO<sub>2</sub> emissions, reduces consumption of energy that in turn leads to 178 179 cheaper products with reduced environmental impact. A brief summary of key experimental research work on this clay processing technique can be found in Table 1. 180

181 El-Mahllawy et al. [36] investigated the effectiveness of combining Kafr Homied clay (KHC), Marble Cutting Waste (MCW), hydrated lime (HL) and Portland cement (PC) in the production 182 of sustainable unfired clay brick. The results indicated that the water absorption decreased 183 184 while the bulk density and compressive strength increased with extended curing time and increase of the MCW content in the presence of HL. This was mainly due to the pozzolanic 185 reaction between KHC, HL and MCW that leads to the formation of cement phases that reduced 186 187 the numbers of pores, decreased the water absorption and evidenced the densification of the mixtures. The X-ray powder diffraction (XRD) results indicated a reduction in the peaks of 188 clay minerals and quartz intensity with increasing MCW content and curing time. This was 189

attributed to the progress of the pozzolanic reaction due to both the high alkalinity environment
and the reaction between silica from the clay and lime from both HL and MCW that led to the
formation of C-S-H gel.

Sekhar and Nayak. [37] studied the influence of using Ground Granulated Blast Furnace Slag 193 (GGBS) and cement in the production of compressed stabilized earth blocks (CSEB) made 194 195 from lithomargic clay. Compressive strength and water absorption of CSEB were evaluated after 28 days. The results indicated a reduction in the water absorption and an increase in the 196 compressive strength with increase in the cement dosage and the air cured CSEB provided 197 higher strength relative to water cured CSEB. The improvement in strength and reduction in 198 water absorption with increase in the cement content was attributed to the formation of 199 additional hydration products that created strong bonds that filled the pores of the soil matrix 200 201 and connected the soil particles in an enhanced structure.

Sitton et al., [38] studied the effect of stabiliser (cement) content and soil to sand ratio (SSR) 202 on the mechanical performance of compressed earth blocks (CSBs) made from silty-clayey 203 soil. Flexural and compressive strength tests after 7 and 28 days of curing either in water or in 204 air were employed to evaluate the performance of the CSBs. The results indicated that strength 205 206 of the CEBs increased with increasing the cement content because the cement has better binding properties than the clay on its own. Additionally, the CSBs under water curing 207 208 exhibited higher strength than that under air curing. This was attributed to the fact that more hydration products from the reaction between the cement and soil could be formed in the 209 presence of water over extended periods. The highest compressive and flexural strengths of 210 15.15 MPa and 1.84 MPa were achieved at cement content of 10.91%, SSR of 3.36 and water 211 content of 11.4% after 28 days under water curing, respectively. 212

Saidi et al. [39] evaluated the effect of cement and lime content on the thermal conductivity of 213 stabilised earth blocks (SEB). The results indicated that with increased stabiliser content, the 214 thermal conductivity increased, which in turn resulted in decreasing the thermal insulation. The 215 results also indicated that lime SEB exhibited lower thermal conductivity relative to cement 216 SEB at the same levels of stabilisation. The increase in cement and lime content in SEB resulted 217 in an increase in the thermal conductivity in comparison with un-stabilised blocks. The increase 218 219 in thermal conductivity of SEB with increasing stabiliser content was attributed to the formation of additional hydration products that filled spaces between the soil particles and 220 221 produced a denser structure.

222 Nshimiyimana et al. [40] carried out experimental work to investigated the effect of using Calcium Carbide Residue (CCR) and Rice Husk Ash (RHA) in the manufacturing of 223 224 compressed earth blocks (CEBs) made from reddish clayey soil. Compressive strength test after 45 days was employed to evaluate the performance of the CEBs. The results indicated 225 that the highest compressive strength of 3.4 MPa was achieved with CCR content of 8% that 226 was about 180% the compressive strength of the control CEBs (100% reddish clayey soil). 227 Additionally, the compressive strengths were significantly improved by the addition of RHA 228 229 along with the CCR. The highest compressive strength of 6.6 MPa was achieved at replacement 230 level of 15% (10.5% CCR and 4.5% RHA) that was almost 3.5 times the compressive strength 231 of the control CEBs. The improvement in the compressive strength in the presence of CCR and RHA is believed to be due to the increased hydration products formed from the reaction of 232 calcium from the CCR and the silica from the RHA. 233

Espuelas et al. [41] investigated the use of magnesium oxide (MgO) rich kiln dust (PC-8) as a binder for the production of unfired clay bricks made from Spanish clay soil. The performance of specimens was assessed by measuring the unconfined compressive strength at 1, 7, 28, 56 and 90 days of curing and water absorption after immersion in water for 24h. The results 238 indicated that the compressive strength increased and water absorption decreased with both (i) the curing time and (ii) increased PC-8 dosage. The results indicated that the optimum dosage 239 240 of PC-8 was 15% that provided a compressive strength of 9.9MPa and a water absorption value of about 5% after 90 days of curing. The development of strength and durability aspects of the 241 developed unfired bricks was attributed to the ability of MgO rich kiln dust binder to form 242 cementitious gels that bind clay soils. Therefore, the results obtained confirmed the suitability 243 244 of MgO based binders as alternative binders to cement or lime in the production of unfired bricks. 245

Abdullah et al. [42] examined the influence of different compactions (14MPa, 21MPa and 246 28MPa) on the performance of Compressed Stabilised Earth Bricks (CSEBs) made from either 247 Laterite Soil, sand and cement or clay, sand and cement. For the evaluation of the performance 248 249 of the compacted brick, compressive strength testing was performed after 7 and 28 days along with water absorption tests. The effect of compaction on compressive strength for clay and 250 laterite soil was contradictory as the optimum strength was achieved by the samples subjected 251 to compaction of 14MPa and 28MPa for laterite soil and clay, respectively. Water absorption 252 testing found that water absorption was improved through increased compaction resulting in 253 254 denser samples with less voids.

Zhang et al. [43] studied the effect of cement content and bulk density on the thermal 255 conductivity and compressive strength of cement stabilised earth blocks (CSEB). The results 256 indicated that the cement content caused only small variations in the thermal conductivity of 257 the CSEB. This was mainly due to the small dosage of cement that had been added to the 258 CSEB, so that it was not sufficient to cause a considerable effect on the thermal conductivity. 259 However, the results of the compressive strength testing indicated a significant improvement 260 in the strength of CSEB by increasing the cement dosage. The results also indicated that by 261 increasing the bulk density, the thermal conductivity and compressive strength of the CSEB 262

increased. This is because increasing the bulk density caused a reduction in the number of poresand decreased the pore diameters in the CSEB significantly.

Taallah and Guettala, 2016 [44] investigated the production of compressed stabilised earth 265 blocks (CSBs) made from Biskra soil and three different percentages of quicklime. 266 Compressive strength and tensile strength were used to evaluate the performance of the CSBs 267 268 after 28 days of either water or air curing. The results indicated higher compressive and tensile strengths of the CSBs under air curing relative to water curing. The results also showed an 269 increased strength with increasing the quicklime content until the optimum dosage of 10%, 270 however, behind this level the strength tend to decrease. The improvement in the strength with 271 increasing the quicklime content was attributed to the formation of additional C-S-H gel. On 272 the other hand, the reduction in the strength of CSBs made with more than 10% quicklime 273 274 content was attributed to the excessive contents of calcite and portlandite  $(Ca(OH)_2)$  with increasing quicklime content that resulted in reduced strength. 275

276 Rahmat et al. [45] investigated the development of unfired brick made with Lower Oxford Clay (LOC) and pulverised fuel ash (PFA). In this study, four stabilisers were used: Lime (L), 277 Portland Cement (PC), lime-GGBS (30:70) and PC-GGBS (40:60). The investigation included 278 279 unconfined compressive strength, water absorption, thermal conductivity and freeze and thaw tests alongside an evaluation of the environmental performance. The results of compressive 280 281 strength and water absorption tests indicated that blending GGBS with lime or PC provided better performance than using only PC or Lime. This was mainly due to the combined 282 pozzolanic reaction that leads to the formation of additional C-A-S-H gel that fills the voids, 283 thus enhancing the strength and reducing the porosity of the brick to a minimum. In addition, 284 the results of thermal conductivity testing indicated that bricks stabilised with PC-GGBS 285 provided the lowest thermal conductivity value. Moreover, the results of the freezing and 286 thawing at the end of the 30th cycle indicated that the weight loss of the bricks increased with 287

increasing the freezing and thawing cycles and the bricks stabilised with PC-GGBS achieved
the lowest percentage of weight loss. Finally, the results of the environmental performance
review suggested that the developed brick can be considered to be green brick with low energy
usage and CO2 emissions and is suitable for the construction of internal walls.

Oti et al. [46] studied the possibility of combining Brick Dust Waste (BDW) from the cutting 292 293 of fired clay bricks with Mercia Mudstone Clay (MMC) in the development of unfired clay (mortar, block and brick). The results indicated an increase in compressive strength with 294 increased percentages of BDW in the mixtures. This was mainly due to the pozzolanic reaction 295 of GGBS and lime that led to the formation of additional C–S–H gel within the pore structure. 296 297 The results also showed that the water absorption rate for all the mixtures was extremely low. Additionally, the results of the weight loss of samples after 7, 28, 56 and 100 cycles of freezing 298 and thawing indicated an increase in the weight loss with increasing BDW content. Overall, 299 the results indicated the potential production of unfired clay products using up to 20% BDW 300 as replacement to MMC with acceptable performance of stabilised clay masonry units. 301

Nagaraj et al. [35] studied the effect of combining cement and lime on the long-term properties 302 of compressed stabilised earth blocks (CSEBs) prepared from red earth. For evaluating the 303 engendering properties of CSEBs, compressive strength and water absorption tests were 304 conducted after 7, 15, 30, 60, 120, 180 days; 1, 2 and 5 years. The results indicated that the 305 306 compressive strength increased and water absorption decreased with increasing the age of curing for all the mixtures. The results also showed that up to the age of 120 days, the strength 307 of the CSEBs stabilised with cement alone was higher than that stabilised with cement and 308 lime. This was attributed to the quick hydration of cement relative to lime that helps the 309 formation of hydration product within the CSEBs. At the age of 180 days onward, the CSEBs 310 311 stabilised with 4% cement and 4% lime provided better strength than that stabilised with 8% cement. After 5 years of curing, the CSEBs with 4% cement and 4% lime have showed a 312

313 compressive strength of 7.2 MPa that was about 167% the strength of CSEBs stabilised with 314 cement alone (4.3 MPa). This behaviour was attributed to the availability of adequate quantity 315 of lime that possibly resulted in increasing the pH of the system and allow the alumina and 316 silica in the clay to be dissolved and to combine with Ca<sup>++</sup> to form calcium-alumino silicates 317 (C-A-S) and thereby binds the particles of clay existing in the matrix.

Miqueleiz et al. [47] evaluated the development of unfired clay brick by blending marl clay 318 soil and alumina filler (AF) waste. The stabilizers used in this investigation were a combination 319 of Pulverised fuel ash (PFA) and lime (L) (70% PFA: 30% L). All mixtures were tested for 320 compressive strength, water absorption and underwent 45 freeze/thaw cycles. The results of 321 the compressive strength testing indicated that increasing the level of AF caused a significant 322 reduction in the compressive strength relative to mixtures made with marl clay soil only at all 323 324 curing ages. The results of water absorption tests showed that the presence of up to 40% AF have a water absorption rate of less than 20%, however the bricks with 60% AF collapsed upon 325 immersion in water at all curing ages. This was attributed to the lower percentages of silica 326 provided by marl clay that combined with calcium from the lime and results in the formation 327 of less hydration products. The results of durability tests indicated that all mixtures were able 328 329 to withstand the repeated 48-hour freezing/thawing cycles without any surface cracks.

| Reference | Clay type         | Blended        | materials         | Clay-based   | Curing condition    | Tests             |
|-----------|-------------------|----------------|-------------------|--------------|---------------------|-------------------|
|           |                   | Replacement    | Stabilising       | product      |                     | conducted         |
|           |                   | material       | material          |              |                     |                   |
| [36]      | Kafr Homied clay  | Marble Cutting | Hydrated lime     | Brick        | Cured in a          | Compressive       |
|           |                   | Waste (0%,     | (0%, 10%, 15%     |              | humidity chamber    | strength, water   |
|           |                   | 10%, 15% and   | and 20%) and      |              | at 40°C±2 for 14    | absorption, bulk  |
|           |                   | 20%)           | 5% Portland       |              | and 28 days         | density, and      |
|           |                   |                | cement            |              |                     | XRD               |
| [37]      | Lithomargic clay  | GGBS 25%       | Cement (0, 6, 8,  | Compressed   | Cured in water and  | Compressive       |
|           |                   |                | 10 and 12 %)      | stabilised   | air for 28 days     | strength and      |
|           |                   |                |                   | earth blocks |                     | water absorption  |
| [38]      | Silty-clayey soil | -              | Cement (3.6,      | Compressed   | Cured in water and  | Compressive       |
|           |                   |                | 5.5, 9.1 and      | stabilised   | air for 7 and 28    | strength and      |
|           |                   |                | 10.9 %)           | earth blocks | days                | flexural strength |
| [39]      | Sidi Amor soil    | -              | Cement (0, 5, 8,  | Stabilised   | Cured in a humid    | Thermal           |
|           |                   |                | 10 and 12%) or    | earth blocks | atmosphere for 28   | conductivity      |
|           |                   |                | lime (0, 5, 8, 10 |              | days at             |                   |
|           |                   |                | and 12%)          |              | temperature of      |                   |
|           |                   |                |                   |              | $20\pm2^{\circ}C$   |                   |
| [40]      | Reddish clayey    | -              | Calcium           | Compressed   | Cured in ambient    | Compressive       |
|           | soil              |                | carbide residue   | stabilised   | condition (30 $\pm$ | strength and      |
|           |                   |                | (0, 5, 8, 10 and  | earth blocks | 5°C) for 45 days.   | SEM               |
|           |                   |                | 15 %) or          |              |                     |                   |
|           |                   |                | calcium carbide   |              |                     |                   |
|           |                   |                | residue and rice  |              |                     |                   |
|           |                   |                | husk ash (0, 5,   |              |                     |                   |
|           |                   |                | 8, 10 and 15 %)   |              |                     |                   |

| [41] | Spanish clay soil         | -                               | Magnesium<br>oxide rich kiln<br>dust (0, 3, 6, 9,<br>12, 15 and<br>18%) | Brick                                    | Cured in wet<br>chamber  | Compressive<br>strength and<br>water absorption  |
|------|---------------------------|---------------------------------|---|--|--|--|
| [42] | Laterite Soil or<br>clay  | Building sand<br>(20%) or (50%) | Cement (10%)<br>or (20%)  | Compressed<br>Stabilised<br>earth brick  | Spraying the<br>samples with<br>water every day up<br>to 7 or 28 days.   | Compressive<br>strength and<br>water absorption  |
| [43] | Local soil in<br>Xinjiang | -                               | Cement (3, 5, 7<br>and 9 %)   | Stabilized<br>earth blocks               | Samples wrapped<br>with plastic foils<br>and placed in the<br>laboratory for 28<br>days at<br>temperature of 20<br>$\pm 1^{\circ}$ C | Compressive<br>strength and<br>thermal<br>conductivity   |
| [44] | Biskra soil               | -                               | Quicklime (8, 10 and 12%)   | Compressed<br>stabilised<br>earth blocks | Cured in water and air for 28 days   | Compressive<br>strength and<br>tensile strengths.  |
| [45] | Lower Oxford<br>Clay      | PFA (50%)                       | Lime-GGBS<br>(30:70) and<br>cement-GGBS<br>(40:60) (10%)                | Brick                                    | Curing in humidity<br>chamber at 20°C<br>for 7 and 28 days   | Compressive<br>Strength, Water<br>Absorption,<br>thermal<br>conductivity<br>freeze and thaw,<br>and<br>environmental<br>performance. |

| [46] | Mercia mudstone | Brick dust     | GGBS and lime | Mortar,      | The samples were    | Compressive      |
|------|-----------------|----------------|---------------|--------------|---------------------|------------------|
|      | clay            | waste (5%,     | (22%)         | block and    | moist-cured for 3,  | Strength, water  |
|      |                 | 10%, 15% and   |               | brick        | 7, 14, 28           | absorption and   |
|      |                 | 20%)           |               |              | and 56 days at      | freeze and thaw  |
|      |                 |                |               |              | room temperature    |                  |
|      |                 |                |               |              | of about 20°C.      |                  |
| [35] | Red soil        | -              | Cement (4, 6  | Compressed   | Cured in water for  | Compressive      |
|      |                 |                | and 8%) and   | stabilised   | 7, 15, 30, 60, 120, | strength and     |
|      |                 |                | lime (0%, 2%  | earth blocks | 180 days; 1, 2 and  | water absorption |
|      |                 |                | and 4%)       |              | 5 years             |                  |
| [47] | Marl clay soil  | Alumina filler | PFA-Lime (70: | Brick        | Cured in a          | Compressive      |
|      |                 | (0, 20, 40 and | 30) (12%)     |              | moisture            | Strength, water  |
|      |                 | 60 %)          |               |              | chamber for 1, 7,   | absorption and   |
|      |                 |                |               |              | 28, 56 and 90 days  | freeze and thaw  |

# 334 2.2.1 Critical evaluation

The technique of producing clay-based building materials by blending and stabilising clay is considered an important step in the fabrication of different clay-based construction materials. The main aim of this technique (that depends mainly on the production of hydration products such as C–S–H and C–A–S–H phases) is enhancing the mechanical and durability performance of the produced product.

According to the reviewed studies and amongst the variety of stabilizers utilised, cement and lime have been the most popular stabilizers in the production of stabilised clay-based construction materials. Cement and/or lime were used either alone [37-39, 42-44], in combination with each other [35, 36] or in combination with other stabilisers such as GGBS or PFA [45-47]. However, two of the reviewed studies were used different stabilisers such as calcium carbide residue with rice husk ash [40] and magnesium oxide rich kiln dust [41].

The dosage of the stabilisers in the reviewed studies varied depending on the type of stabiliser. 346 347 For example, the cement dosage was ranging between 3% and 12% [37-39, 43] and only one paper [42] have showed the usage of 20% cement as stabiliser. The dosage of lime as stabiliser 348 was similar to that of the cement (ranging between 2% and 12%). Additionally, the lime and 349 cement were blended with GGBS or PFA to boost the hydration process of these materials by 350 the highly alkaline environment (pH > 12) and accelerate the production of cementitious 351 compound that binds the soil together [45, 46]. In some cases, the stabiliser dosage were 352 relatively high with 22% for [46] and 25% for [36]. Regarding the dosage of stabiliser for the 353 paper with calcium carbide residue and rice husk ash [40] were in the range of 5-12% while 354 the magnesium oxide rich kiln dust [41] was in the range of 3-18%. 355

The reviewed studies indicated that the production of C–S–H and C–A–S–H gel phases increased with increasing the amounts of stabiliser in the mixture [36, 37, 39, 40, 45]. This was

| 358 | due to the chemical reaction occurring between the CaO from the cement, lime and other            |
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| 359 | stabilisers and amorphous silica provided by clay together with the high alkalinity environment   |
| 360 | of cement or lime [37, 39, 45, 46]. These gels tend to fill pores and grow into capillary spaces, |
| 361 | resulting in a more impermeable, dense and higher-strength structure [48, 49]. However, high      |
| 362 | dosage of stabiliser with high CaO content might negatively affect the performance of the         |
| 363 | product because this might resulted in free CaO that could lead to expansion and cracks in the    |
| 364 | final product [9].  |
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# 378 2.3 Production of geopolymer clay-based construction materials through alkali activation 379 (Geopolymerisation)

380 During the development of construction materials using the technique of geopolymerisation, the most important chemical compounds that should be available in high a quantity are the SiO<sub>2</sub> 381 and Al<sub>2</sub>O<sub>3</sub> because they react with a high alkaline solution to produce amorphous to semi-382 383 crystalline geopolymer material [5, 17]. Preferably, the total composition of the SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> compounds should be more than 70% [17]. Yun-Ming et al. [17] stated that the raw materials 384 with abundantly amount of  $SiO_2$  and  $Al_2O_3$  that is suitable for the production of geopolymer 385 materials can be found in different types of clay. These observations are in consistent with the 386 results obtained from Fig.1 for different types of clay that were used in the production of 387 geopoylmer clay-based construction materials. 388

This technique is of great research interest in the field of sustainable construction materials due to its characteristics such as developing high mechanical strength within a very short time, enhanced durability, high fire resistance and considerably decreased greenhouse gas emissions and energy consumption [23, 50, 51]. Many researchers have investigated the production of clay-based construction materials using the technique of alkali activation (geopolymerisation) (see Table 2).

Faqir et al. [52] conducted experimental work to investigate the utilisation of kaolin clay activated with NaOH in different concentrations in the fabrication of geopolymer mortar. In this study, the effect of sand to kaolin clay ratio and NaOH with different concentrations to kaolin clay ratio were evaluated by determine the compressive strength of geopoymer samples after 7 days of water and air curing. The results indicated that with decreasing the sand to kaolin clay ratio and increasing both the concentration and the amount of the NaOH, the compressive strength increased for both curing methods. The highest compressive strength were 27.1 MPa 402 and 18.1 MPa achieved at sand to kaolin clay ratio of 1.5 and NaOH (with concentration of 17 M) to kaolin clay ratio of 0.17 under air curing and water curing, respectively. Additionally, 403 404 the results of the water absorption test indicated that the geopolymer mortar exhibits lower 405 water absorption rate with increasing the concentration of the NaOH and the lowest water absorption was about 3.7 % achieved for the geopolymer mortar that showed the highest 406 compressive strength. The improvement in the strength and durability of the geopolymer 407 408 mortar with increasing the NaOH concentration is believed to be due to the formation of denser microstructure that bonded the kaolin clay and the sand with less numbers of pores. 409

Dassekpo et al. [53] evaluated the use of clay waste (CW) from construction sites, class F fly 410 ash (FA), sodium hydroxide (NaOH) and sodium silicate (Na2SiO3) in the production of 411 geopolymer paste. The experimental programme included evaluation compressive strength, 412 Scanning Electron Microscopy (SEM), Energy Dispersive X-Ray Spectroscopy (EDX) and the 413 leaching behaviour of the geopolymer paste. The results indicated an increase in the 414 compressive strength with increases in the curing period and the FA content. This was due to 415 the enhanced polymerisation process with longer curing periods and the increased Si/Al ratio 416 with increased content of FA, which in turn resulted in increasing the amount of Si-O-Si bonds 417 that formed during the geopolymerisation process, as evidenced by the EDX test. The SEM 418 419 images showed an increase in the degree of compaction and formation of a denser 420 microstructure as the amount of FA in the pastes increased. The leaching test was conducted for samples soaked in deionized water for 4, 8, 12, 24, 72 and 336 hours to measure the 421 concentration of Aluminium (Al) and Arsenic (As). The results indicated that with increasing 422 the FA content, the Al concentration decreased and As concentration increased. 423

Sore et al. [51] investigated the development of geopolymer compressed earth blocks (CEB)
using laterite clay, metakaolin (MK) and sodium hydroxide. The experimental programme
included weight loss after curing, porosity, apparent density, compressive strength, flexural

427 strength, thermal diffusivity and thermal conductivity. The results indicated that with increasing MK content, the weight loss and porosity increased, while the apparent density 428 decreased. In addition, the results of compressive and flexural strength tests revealed that with 429 430 increased MK content, the strength of geopolymer CEB improved. This improvement in the mechanical strength was attributed to the formation of higher levels of geopolymer gels after a 431 polymerisation reaction between the NaOH and the MK that resulted in increasing the bond of 432 433 particles, resulting in a more resistant and more compact structure. Furthermore, the results also indicated low thermal diffusivity and thermal conductivity with all MK percentages. 434

Phummiphan et al. [22] studied the development of low carbon pavement base material 435 produced from lateritic soil (LS), class C FA, Granulated Blast Furnace Slag (GBFS), sodium 436 hydroxide and sodium silicate. The performance of the low carbon pavement base material was 437 438 evaluated in terms of unconfined compressive strength, SEM and XRD tests after 7, 28 and 60 days. The results indicated that the highest compressive strength values after 28 and 60 days 439 were found at LS: FA: GBFS = 60:30:10 and Na2SiO3: NaOH of 90:10. This mixture was 440 considered to be the recommended optimum ratio in practice and was further investigated for 441 SEM and XRD tests. The results of SEM and XRD tests indicated that the geopolymerisation 442 products increased in volume as the curing time increased. 443

Miranda et al. [54] investigated the development of low carbon alkali activated mortar (AAM) produced from granitic residual soil (GRS), FA, sodium silicate and sodium hydroxide. The mechanical performance of the AAM were evaluated by means of compressive strength and flexural strength after 30, 60 and 90 days of curing. The results indicated an improvement in the compressive and flexural strengths with increasing the curing time and FA content. In addition, the performance of the developed mortars was also assessed by building masonry walls using compressed earth blocks utilising the AAM. The results showed that walls 451 incorporating AAM with 15% FA had better performance than walls incorporating AAM with452 5% FA, but the increment was minimal.

453 Leitão et al. [55] evaluated the mechanical and thermal performances of Alkali Activated Interlocking Compressed Earth Blocks (AAICEBs) made from 85% granitic residual soil, 15% 454 FA, sodium hydroxide and sodium silicate. For assessing the mechanical performance of 455 456 AAICEBs, the compressive strength test was conducted on the AAICEBs after 28 days. Additionally, the developed AAICEBs were used after 28 days in the building of a masonry 457 wall and then the thermal conductivity of that wall was evaluated. The results indicated a 458 compressive strength of 3MPa after 28 days of curing. The results of the thermal conductivity 459 of the masonry wall suggested that the use of alkaline activators in the presence of 15% FA 460 improved the thermal properties of the wall with respect to heat transference. 461

462 Messina et al. [56] investigated the utilisation of calcined clay sediments (CCS) and calcined water potabilization sludge (CWPS) in the production of precast geopolymer paying elements. 463 464 The raw materials were calcined at 750°C for two hours, and then different proportions of the calcined raw materials were blended with Na<sub>2</sub>SiO<sub>3</sub> and NaOH to produce geopolymer paste. 465 The developed paste was then mixed with building sand to produce geopolymer mortar. 466 Compressive strength test was used to evaluate the performance of the geopolymer paste and 467 mortar after 7 days of curing. The results indicated that the compressive strength of the 468 469 developed pastes and mortars were very similar and were in the range between 17-23 MPa. The developed mortar with CCS/WPS ratio of 50/50 was then mixed with natural aggregate to 470 produce paving bricks that have been evaluated by measuring the splitting tensile strength after 471 7 days of curing. The developed paving bricks have showed splitting tensile strength between 472 0.82-2.01 MPa. The durability assessment of the developed bricks after 180 days of exposure 473 to room conditions showed that there was no cracking and no efflorescence was formed on the 474 surface of the developed paving brick. 475

Slaty et al. [23] studied the durability performance of geopolymer mortar and paste made from 476 Jordanian Hiswa kaolinite (JHK) clay and sodium hydroxide. The experimental programme 477 included drying shrinkage, wetting and drying conditions, sea water attack and alkali-silica 478 479 reaction. The results indicated a very low drying shrinkage for all samples and the presence of sand in mortar significantly reduced the shrinkage relative to geopolymer paste. The results 480 also showed that there was a 50% reduction in the compressive strength of specimens subjected 481 482 to 100 cycles of wetting and drying conditions relative to geopolymer specimens cured under dry conditions. This was attributed to effect of water in weakening the bond strength of Si-O-483 484 Si in the alumina-silicates resource and because clay minerals have a high tendency to absorb water and become plastic rather than stiff. In addition, the results indicated a very good 485 mechanical performance for geopolymer mortar and paste immersed in sea water with a small 486 487 formation of efflorescent material on the external surface of the samples. Furthermore, the alkali-silica reaction seriously affected the geopolymer specimens and produced expansion, 488 cracking, and loss of the mechanical strength as a function of time. 489

Poowancum and Horpibulsuk [57] investigated the use of Dan Kwian sedimentary clay 490 (DKSC) in the development of geopolymer binder. During this investigation, the DKSC was 491 492 calcined at 600°C for 1, 2 and 5 h and mixed with Na<sub>2</sub>SiO<sub>3</sub> solution and NaOH solution in three 493 different Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratios (0.5, 1 and 1.5). Setting time, compressive strength and porosity 494 were used to evaluate the effect of different calcination temperatures and Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratios on the properties of the geopolymer paste. The results indicated that for a fixed Na<sub>2</sub>SiO<sub>3</sub>/NaOH 495 ratio, the setting time of the paste calcined for 1 and 2 h was about 55 min and was about 60 496 min for the paste calcined for 5 h. The increase in the setting time with increasing the 497 498 calcination time was associated with a reduction in the compressive strength and higher porosity of the geopolymer paste. The results also showed that the compressive strength of the 499 geopolymer paste decreased with increasing the Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratio and the lowest 500

501 compressive strength was recorded for Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratio of 1. Beyond this ratio, the 502 compressive strength improved with increasing Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratio. The highest compressive 503 strength of 27 MPa and lowest porosity of 35% was achieved with calcination of the DKSC 504 for 2 h and Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratio of 0.5.

Silva et al. [58] investigated the production of compressed earth blocks (CEBs) from granitic 505 506 residual soil, FA, sodium hydroxide and sodium silicate. Compressive strength and flexural strength after 180 days of curing were used for assessing the performance of the CEBs. The 507 508 results showed superior compressive and flexural strengths of CEBs incorporating 15% FA relative to the CEBs with 10% FA. This was attributed to the formation of more geopolymer 509 products along with the enhanced structure due to higher density. The performance of the CEBs 510 in saturated conditions was also evaluated by submerging CEBs in water for 24 hours prior to 511 512 the test. The results indicated a maximum reduction in the compressive and flexural strengths of 36% and 61%, respectively for the mixture with 10% FA in comparison to CEBs samples 513 514 cured in dry conditions.

Ferone et al. [59] carried out experimental works to investigate the possibility of employing 515 Sabetta clay sediments (SCS) in the production of geopolymer binder. During this 516 investigation, the SCS was calcined at two different temperatures (400°C or 750°C) for 120 517 minutes and activated by NaOH alone or a mixture of NaOH and Na<sub>2</sub>SiO<sub>3</sub>. Compressive 518 strength test was conducted after 28 days to assess the behaviour of the binder under different 519 conditions of calcination and activation. The results indicated that compressive strength of the 520 geopolymer binder was significantly enhanced with increasing the calcination temperature 521 from 400°C to 750°C. Additionally, the results showed that the compressive strength was 522 improved with increasing the concentration of the NaOH and when the SCS was activated by 523 a mixture of NaOH and Na<sub>2</sub>SiO<sub>3</sub>. The compressive strength was further enhanced by the 524 addition of 17 % GGBS. The highest compressive strength value of the geopolymer binder was 525

526 38.9 MPa achieved with SCS treated at 750 °C mixed with 17 % GGBS and activated by a 527 mixture of NaOH and Na<sub>2</sub>SiO<sub>3</sub>. The SEM images showed that the addition of GGBS to the 528 geopolymer binder in the presence of NaOH and Na<sub>2</sub>SiO<sub>3</sub> leads to the formation of a very dense 529 and homogenous microstructure due to the simultaneous formation of N-A-S-H and C-A-S-H 530 gels as evidenced by the EDX test.

531 Phetchuay et al. [60] evaluated the development of pavement subgrade material from silty clay soil, FA, Calcium Carbide Residue (CCR) and sodium silicate. In this investigation, the 532 influential factors were FA replacement level, Na2SiO3/water ratio, curing temperature and 533 curing time for a fixed CCR content of 7%. Compressive strength and SEM tests were used for 534 evaluating the effect of different factors on the strength and microstructure of the pavement 535 subgrade material. The results indicated that the optimum FA replacement level was 15%. 536 Regarding the effect of Na2SiO3/water ratio, the results showed that it was 0.6 for samples 537 under water curing and 1.4 for samples under air curing. Additionally, the results also indicated 538 an increase in the compressive strength with the increase in curing period and curing 539 temperature. The SEM images confirmed the results of compressive strength test that at 540 optimum conditions, a denser microstructure was formed. 541

Molino et al. [61] studied the effect of different calcination temperatures and alkaline activators 542 on the performance of geopolymer binder produced from Occhito clay sediments. During this 543 research, the raw materials were calcined at either 650°C or 750°C for 60 minutes and activated 544 by three alkaline activators, namely NaOH solution, sodium aluminate solution and potassium 545 aluminate solution. Compressive strength test after 3 and 14 days of curing was employed to 546 evaluate the performance of the binder under different conditions of calcination and activation. 547 At the age of 3 days, the results indicated an improvement in the compressive strength of the 548 binder with increasing the temperature of calcination from 650°C to 750°C for the samples 549 activated with either NaOH solution or sodium aluminate solution, while decreased for samples 550

551 activated with potassium aluminate solution. After 14 days of curing, the results indicated higher compressive strength with increasing the temperature of calcination for all the used 552 activators. The SEM images of the samples made with sodium aluminate solution showed a 553 compacted microstructure with no significant changes with increasing the age of curing or the 554 temperature of calcination. In summary, the utilisation of sodium aluminate activator at 555 calcination temperature of 650°C provided higher compressive strength than other tested 556 activators with calcination temperature of 750°C, thus the use of this activator could 557 significantly reduce the energy usage and improve the sustainability of the final product. 558

Slaty et al. [62] investigated the development of alkali-activated mortar using kaolinitic clay, 559 silica sand and sodium hydroxide. The experimental programme included optimising the sand 560 to binder ratio, curing temperature, and curing period. The results showed that by increasing 561 562 the sand content, the workability of mortar improved and the highest compressive strength value was achieved when the sand to clay ratio was 1. The results also indicated that with 563 increasing the curing temperature from 50oC to 80oC, the compressive strength increased from 564 14MPa to 32MPa after 24h of curing. Additionally, the results indicated an increase in 565 compressive strength with increased curing time. This study concluded that the optimum 566 conditions for producing kaolinitic clay-based mortar were; sand to kaolinitic clay ratio of 1, a 567 curing temperature of 80oC and curing time of 24h. Furthermore, the optimised samples were 568 569 tested under wet and dry conditions. The results showed a reduction in compressive strength by half for samples under wet conditions relative to dry conditions. This was attributed to the 570 hydrolysis of the Si-O-Si bonds upon immersion in water. The results of the SEM and XRD 571 tests evidenced the formation of crystalline reaction products that filled the pore spaces and 572 573 helped bind the matrix.

Sukmak et al. [63] examined the development of geopolymer brick using silty clay soil, FA,
sodium hydroxide and sodium silicate. The Na2SiO3 / NaOH ratios studied were 0.4, 0.7, 1.0

576 and 1.5. Additionally, different Liquid (L)/FA ratios (0.4, 0.5, 0.6 and 0.7) and FA/clay ratios (0.3, 0.5 and 0.7) were investigated. The experimental programme included measuring the 577 compressive strength of brick after 7, 14, 28, 60, and 90 days of curing at ambient temperature. 578 579 The results indicated that for different L/FA and FA/clay ratios, the optimum compressive strength was for the mixtures with Na2SiO3/NaOH ratio of 0.7. For a given Na2SiO3/NaOH 580 ratio, the strength increased with increasing L/FA ratio until its optimum value was reached, it 581 582 then tended to decrease. The results also indicated that the compressive strength increased with increases in the FA content. This was due the increased geopolymerisation products produced 583 584 because of the high alumina-silicate of FA. The overall results indicated that the optimum L/FA ratio was dependent upon only the FA/clay ratio. As the clay content decreases, the L required 585 for the reaction decreases. 586

Mohsen and Mostafa [64] studied the use of calcined white clay (CWC) in the development of 587 geopolymer bricks. The clay was calcined at 700°C for two hours and then mixed with either 588 NaOH alone or with a mixture of Na<sub>2</sub>SiO<sub>3</sub> and NaOH to produce geopolymer bricks. For 589 evaluating the performance of the geoploymer bricks, compressive strength test was conducted 590 at (i) room temperature for 3 days, (ii) 75°C for 24 h and (iii) 150°C for 24 h. The results 591 592 indicated that the compressive strength of the developed bricks with NaOH was improved from 19.8 MPa to about 22 MPa with increasing the curing temperature from room temperature to 593 594 75°C. However, increasing the curing temperature to 150°C resulted in a slight reduction in the compressive strength (18 MPa). Additionally, the compressive strength results of the 595 geopolymer bricks activated with Na<sub>2</sub>SiO<sub>3</sub> and NaOH improved from 44 MPa to about 79 MPa 596 with increasing the curing temperature. The results of water absorption test indicated that 597 598 increasing the curing temperature reduced the water absorption of the produced bricks for both activators. In summary, geopolymer bricks activated with Na<sub>2</sub>SiO<sub>3</sub> and NaOH have showed 599 higher compressive strength and much lower water absorption values than those activated with 600

- 601 NaOH solution only. This was attributed to the formation of denser geopolymer gel for bricks
- activated with  $Na_2SiO_3$  and NaOH relative to those activated with NaOH solution only as
- 603 observed by the SEM testing.

**Table 2.** Studies on production of clay-based construction and building materials through alkali activation (geopolymerisation)

| Reference | Clay type                            | Blended material   | Alkali activator                           | Clay-based<br>product      | Curing condition   | Tests conducted  |
|-----------|--------------------------------------|--|--|----------------------------|--|--|
| [52]      | Kaolin clay                          | -  | NaOH (13, 16.3,<br>17, 17.8 and 19.7<br>M) | Mortar                     | Cured in oven at<br>80°C for 24 hours<br>then at air or in<br>water for 7 days   | Compressive<br>strength and water<br>absorption  |
| [53]      | Clay waste from<br>construction site | Class F fly ash (0,<br>10, 20 and 30%)   | Na2SiO3 and<br>NaOH (14M)                  | Paste                      | Cured in a<br>humidity chamber<br>at 75°C±2 for 24<br>hours then at<br>ambient<br>temperature for 7,<br>14 and 28 days   | Compressive<br>strength,<br>SEM/EDX and<br>leaching<br>behaviour   |
| [51]      | Laterite clay                        | Metakaolin (0, 5, 10, 15 and 20%)  | NaOH (12M)                                 | Compressed earth<br>blocks | Cured at ambient<br>temperature of<br>$(30^{\circ}C \pm 5^{\circ}C)$ for 7<br>days and then<br>placed in oven for<br>another 7 days at<br>$60^{\circ}C \pm 2^{\circ}C$ | Weight loss after<br>curing, porosity,<br>apparent density,<br>compressive<br>strength, flexural<br>strength, thermal<br>diffusivity and<br>thermal<br>conductivity. |
| [22]      | Lateritic soil                       | Class C fly ash<br>(30%) and<br>Granulated Blast<br>Furnace Slag (10,<br>20 and 30%) | Na2SiO3 and<br>NaOH (5M)                   | Pavement base<br>material  | Wrapped with<br>plastic sheets and<br>cured at room<br>temperature<br>between (27–<br>30)°C for 7, 28<br>and 60 days   | Compressive<br>Strength, SEM<br>and XRD  |

| [54] | Granitic residual<br>soil         | Fly ash (5% and 15%)  | Na <sub>2</sub> SiO <sub>3</sub> and<br>NaOH (5M and<br>12.5M) | Mortar                         | Cured at ambient<br>temperature for<br>30, 60 and 90<br>days  | Compressive<br>strength and<br>flexural strength  |
|------|-----------------------------------|---|--|--------------------------------|---|---|
| [55] | Granitic residual<br>soil         | Fly ash (15%)   | Na <sub>2</sub> SiO <sub>3</sub> and<br>NaOH (12.5M)           | Compressed earth<br>block      | Cured at ambient<br>temperature for<br>28 days  | Compressive<br>strength and<br>thermal<br>conductivity  |
| [56] | Calcined clay<br>sediments        | Calcined water<br>potabilization<br>sludge (30, 50 and<br>70 %) | Na2SiO3 and<br>NaOH (14 M)                                     | Paste, mortar and paving brick | Cured for 24h or<br>either at 20°C or<br>60°C and then at<br>climatic chamber<br>operating at 20°C<br>until the age of 7<br>days. | Compressive<br>strength, splitting<br>tensile strength,<br>SEM, XRD and<br>visual assessment.   |
| [23] | Jordanian Hiswa<br>kaolinite clay | _   | NaOH   | Mortar and paste               | Cured for 24h or<br>48h at 80 °C and<br>then for 7, 30, 60,<br>90 and 180 days<br>at ambient<br>temperature.                      | Compressive<br>strength, drying<br>shrinkage, wetting<br>and drying<br>conditions, sea<br>water attack and<br>alkali-silica<br>reaction |
| [57] | Dan Kwian<br>sedimentary clay     | -   | Na2SiO3 and<br>NaOH (8 M)                                      | Paste                          | Cured at 60°C for<br>7 days   | Setting time,<br>compressive<br>strength and<br>Porosity.   |
| [58] | Granitic residual<br>soil         | Fly ash (10% and 15%)   | Na <sub>2</sub> SiO <sub>3</sub> and<br>NaOH (12.5M)           | Compressed earth<br>block      | Cured at ambient<br>temperature for<br>180 days   | Compressive<br>strength and<br>flexural strength  |

| [59] | Sabetta clay<br>sediments | GGBS (0 and 17<br>%)   | Na <sub>2</sub> SiO <sub>3</sub> and<br>NaOH (5, 7 and<br>10 M)  | Paste                         | Cured for 3 days<br>at 60°C in an<br>oven, then at<br>room temperature<br>until the age of 28<br>days.  | Compressive<br>strength, SEM,<br>EDX and XRD.                                  |
|------|---------------------------|--|--|-------------------------------|---|--|
| [60] | Silty clay soil           | Fly ash (0, 5, 10,<br>15, and 20 %) and<br>7% Calcium<br>Carbide Residue | Na <sub>2</sub> SiO <sub>3</sub>   | Pavement<br>subgrade material | Cured either at<br>ambient<br>temperature (27–<br>30)°C or at 40°C<br>for 7, 14, 28 and<br>60 days      | Unconfined<br>Compressive<br>Strength and SEM                                  |
| [61] | Occhito clay<br>sediments | -  | NaOH (5 M),<br>sodium aluminate<br>(8.5, 11, 13 and<br>17 M) and<br>potassium<br>aluminate (8.5,<br>11, 13 and 17 M) | Paste                         | Cured for 3 days<br>at 60°C in an<br>oven, then kept in<br>air for 11 days                              | Unconfined<br>Compressive<br>Strength and SEM                                  |
| [62] | Kaolinitic clay           | Silica sand (25,<br>50, 100 and<br>150%)                                 | NaOH   | Mortar                        | Cured at<br>temperature (50,<br>60, 70 and 80°C)<br>for curing period<br>(6, 12, 18, 24, 48<br>and 72h) | Workability,<br>compressive<br>strength, wetting<br>and drying, SEM<br>and XRD |
| [63] | Silty clay soil           | Fly ash (30, 50<br>and 70%)  | Na <sub>2</sub> SiO <sub>3</sub> and<br>NaOH (10M)   | Brick                         | Cured at ambient<br>temperature for 7,<br>14, 28, 60, and 90<br>days                                    | Compressive<br>strength  |

|     | [64] | White clay | - | NaOH alone or<br>Na2SiO3 and<br>NaOH | Brick | Curried at (i)<br>room temperature<br>for 3 days, (ii)<br>75°C for 24 h and<br>(iii) 150°C for 24<br>h | Compressive<br>strength, Water<br>absorption, SEM<br>and XRD. |
|-----|------|------------|---|--------------------------------------|-------|--|---|
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| 616 |      |            |   |                                      |       |  |   |
| 617 |      |            |   |                                      |       |  |   |

# 618 2.3.1 Critical evaluation

In some of the reviewed papers, the clay materials were blended with different materials such as FA [53-55, 58, 60, 63], GGBS [59], metakaolin [51] and calcined water potabilization sludge [56]. The main reason behind blending the clay with the aforementioned materials is to increase the geopolymerisation products that in turns resulted in better mechanical and durability performance [17].

624 In the reviewed studies, both class C and Class F fly ash types were used. The reaction between alkaline activator and class C fly ash forms in addition to the geopolymerisation products that 625 626 normally formed during the activation of class F fly ash, Calcium Silicate Hydrate (C-S-H) gel and Calcium Alumino Hydrate (C-A-H) gel [22]. This behaviour is similar to the alkali 627 activation of GGBS and is attributed to the adequate calcium content in class C fly ash and 628 629 GGBS [21, 65]. The amount of fly ash blended with different types of clay that has high SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> content was in the range of 5-30% [22, 53-55, 58, 60]. However, the amount of 630 blended fly ash reached about 70% in the case of clay with total SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> content of 631 about 28% [63]. 632

According to the reviewed studies, the most commonly used alkaline activator solutions were 633 sodium hydroxide (NaOH) and sodium silicate (Na<sub>2</sub>SiO<sub>3</sub>). Sodium hydroxide was used alone 634 as an activator [23, 51, 52, 62] or in combination with sodium silicate [22, 53-59, 63, 64]. The 635 636 utilisation of both activators together in the production of clay-based geopolymer products was vital because NaOH is required for the dissolution of alumina-silicate precursor, while Na<sub>2</sub>SiO<sub>3</sub> 637 acts as binder or alkali reactant [17, 66-68]. Therefore, the final product will have better 638 mechanical and durability performance [59, 64]. Additionally, the use of a combination of 639 640 NaOH and Na<sub>2</sub>SiO<sub>3</sub> is cost effective to produce clay-based geopolymer materials with good compressive strength and durability performance because NaOH is cheaper than Na<sub>2</sub>SiO<sub>3</sub> [17, 641 642 57].

In many of the reviewed papers that utilised NaOH with different concentration (in the range of 5-19.7 M) [52, 59], the results indicated an improvement in the strength and reduction in the water absorption with increasing the NaOH concentration. This enhancement in the strength and durability perfomace of the geopolymer materials is believed to be due to the formation of denser microstructure that bonded the particles of the raw materials, thus resulted in less numbers of pores [17, 52].

Generally, the Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratio in the preparation of clay-based geopolymer products is 649 important [17, 68]. Based on the reviewed studies the Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratios were in the range 650 of 0.25 to 9. The optimum Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratio in the reviewed studies varied significantly 651 according to the type of clay, blended materials, methods of curing, etc. In general, increasing 652 the Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratio resulted in improved strength and durability of the final product. This 653 654 could be attributed to the increased (Si) content that aids in the production of more of Si-O-Si bonds, and significantly enhanced the compressive strength of the clay-based geopolymer 655 materials [63, 69, 70]. However, the strength started to decrease behind the optimum 656 Na<sub>2</sub>SiO<sub>3</sub>/NaOH ratio. This could be due to the excessed Si content that hinders water 657 evaporation and structure formation and negatively affect the geopolymerisation rate [63, 69, 658 659 70].

In addition to the aforementioned alkaline activators, sodium aluminate and potassium aluminate activators were used by Molino et al. [61] with the aim of improving the performance of the raw materials with low  $Al_2O_3$  content (16.33%). The results indicated an improvement in the performance of geopolymer binder with the use of alkaline aluminate solutions relative to NaOH solution.

In some of the reviewed geopolymer-related studies, the raw materials (clays) were calcined at different temperatures and times [56, 57, 59, 61, 64], while the other studies used non-calcined

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clays. Ferone et al., 2015 [59] stated that calcined clay could provide better performance than non-calcined clay. This is because heat treatment helps to transform the crystalline phases into reactive amorphous of raw materials that leads to enhance the strength of geopolymers [17, 59, 71]. Additionally, the reviewed studies showed that the temperature at which the clays were calcined were in the range of 400°C to 750°C while the calcination time was between 1-5 hours. Generally, the strength of different calcined clay-based construction materials has improved with increasing the temperatures of calcination [59, 64] and the optimum calcination time was 2 hours [56, 57, 59, 64]. The improvement in the strength with increasing the calcination temperature is believed to be due to the increased surface area of raw materials that dissolves quicker in the alkaline solution and consequent improve the geopolymerisation reaction [17]. On the other hand, the reduction in strength with increasing the time of calcination for more than two hours is attributed to the over calcination that leads in the transformation of reactive amorphous phase into mullite crystalline phases that are dead burnt and not reactive [17]. 

#### 689 2.4 Microwave sintering, curing and drying of clay-based construction materials.

Microwaves are electromagnetic radiation with frequencies in the range of 300GHz -- 300MHz 690 and wavelengths between 1mm and 1m in free space [31, 72]. Standard microwave for 691 scientific or industrial processing operates at a frequency of 2.45GHz [29, 31, 72, 73]. 692 Sintering, curing and drying of clay-based building materials via microwave technique depends 693 694 mainly on the deep penetration with uniform volumetric heating that significantly reduces processing times due to the rapid heating rate [74-76]. Although many studies have been carried 695 out on the use of microwave techniques in the processing of construction materials [30, 34, 74-696 82], only limited research has been conducted on clay-based construction materials as shown 697 in Table 3. 698

699 Hájková [83] investigated the utilisation of microwave technique to accelerate the curing of 700 geopolymer mortar based on calcined kaolinite claystone (CKC). The geopolymer mortar was manufactured my mixing CKC (calcined at temperature of 750°C), sand and potassium water 701 702 glass. The variables investigated during this study were the density of the potassium water glass (1.2, 1.3, 1.4, 1.5 and 1.6 g/cm<sup>3</sup>) used in the preparation of the geopolymer mortar and the 703 method of microwave curing. The microwave curing methods employed were (1) the 704 705 application of microwave for 26 min immediately after casting the geopolemer mortar (MW) and (2) the application of microwave for 26 min after 24 hours of casting the samples and 706 707 solidification at room temperature (MW/2). Compressive strength test after 7 and 28 days, leaching test of the Si, Al, K and Na elements after 28 days and porosity test were used to 708 evaluate the performance of the geopolymer mortar. The results indicated that the compressive 709 strength was increased with increasing the potassium water glass densities for both curing 710 methods. Additionally, the compressive strengths for all the tested densities and for both curing 711 methods were almost the same at the age of 7 and 28 days. This could be attributed to the fast 712 solidification of the geopolymer mortars under microwave curing that force the polymerization 713

714 process to stop quickly that resulted in similar strengths. The leaching test results indicated an increasing in the leaching of Si, K and Na and reduction in the leaching of Al with increasing 715 the potassium water glass densities. Moreover, the porosity test result indicated that the 716 717 porosity decreased with increasing the potassium water glass densities and the samples with MW curing have higher pore volumes and pore diameters relative to that with MW/2 curing. 718 In summary, the results showed the probable production of geopolymer mortar with 719 720 compressive strength of more than 60 MPa from CKC at the optimum microwave curing conditions detailed in Table 3. 721

722 Taurino et al. [24] investigated the feasibility of using microwave sintering in the production of brick from kaolin clay and municipal solid waste incineration bottom ash (BA). Sintering 723 experiments were carried out at a power rating of 950W for 5 minutes holding at three different 724 725 temperatures (800, 900 and 1000°C). Compressive strength, linear shrinkage and water absorption tests were used to evaluate the performance of the newly developed brick. The 726 results indicated that by increasing the temperature, the liner shrinkage and water absorption 727 728 reduced, while the compressive strength improved. This study concluded that the microwave 729 sintering at optimum microwave conditions detailed in Table 3 for the mix with 55% BA with 730 45% kaolin clay was sufficient to produce brick with 65MPa strength after 28 days of curing,

Bagaber and Sudin, [25] examined the effectiveness of the microwave technique in drying of 731 732 clay bricks as an alternative method to oven drying. In this study, the brick was made from 97% red clay and 3% charcoal. The main reason for adding a small percentage of charcoal was 733 to enhance microwave absorption. The performance of dried brick using the microwave 734 technique was evaluated by measuring density, cracks and water absorption with comparisons 735 made with those dried in a conventional electrical oven. Dried clay bricks using the microwave 736 technique showed improved density, less water absorption and were free from cracks as 737 compared with drying in a conventional electrical oven. In addition, using the microwave 738

technique at optimum conditions, as identified in Table 3, significantly reduced the temperatureand time of drying compared to conventional electrical oven treatment.

Kim et al. [26] evaluated the employment of the microwave technique to accelerate the curing 741 of alkali activated Hwangtoh clay (AAHC) Paste. In this investigation, the alkali activator was 742 a combination of Na2SiO3 and NaOH. Internal temperature distribution, porosity and 743 744 compressive strength tests were used for evaluating the performance of the AAHC paste. For comparison purposes, there were some samples cured using heat curing at 60oC. The results of 745 746 the internal temperature distribution indicated that the core temperature was generally higher than the surface temperature. However, the maximum difference between core and surface was 747 less than 10oC, this evidenced the uniform heating of the microwave technique. The results 748 also showed a reduction in the cumulative pore volume and improvement in the compressive 749 750 strength of AAHC paste with increased microwave curing time. This was due to the gradual filling of larger pores with the reaction products of alkali activation. Additionally, the results 751 752 revealed the possible production of paste with compressive strength of about 21MPa at the optimum microwave curing conditions detailed in Table 3. These results were higher than those 753 754 achieved with conventional heat curing at 60oC for 72 hours.

755 Itaya et al. [27] studied the possibility of using microwave techniques in the drying of kaolin clay bricks as an alternative to conventional oven drying. Deformation and formation of cracks 756 within the brick were used to assess the effectiveness of the microwave drying technique. The 757 results indicated that microwave drying of bricks with constant power resulted in large cracks 758 and breaking of samples when the internal temperature reached about 100oC. However, the 759 results showed successful drying without any deformation or crack formation when the drying 760 process was conducted at optimum drying conditions stated in Table 3. In addition, the use of 761 optimum drying conditions significantly reduced the drying time of kaolin clay brick relative 762 to conventional oven drying 763

**Table 3**. Studies on the use of microwave as sintering, curing and drying technique of clay-based construction materials.

| Reference | Clay type              | Microwave<br>Process | Microwave Power<br>(W)                                     | Microwave<br>Time (minutes)                 | Clay-based<br>product | Tests conducted   | Optimum<br>Microwave<br>Conditions  |
|-----------|------------------------|----------------------|--|---|-----------------------|---|---|
| [83]      | Kaolinite<br>claystone | Curing               | 3000   | 26  | Mortar                | Compressive<br>strength, leaching<br>and porosity                                   | The application of<br>microwave for 26<br>min with power of<br>3000 W after 24<br>hours of casting the<br>samples |
| [24]      | Kaolin clay            | Sintering            | 950  | 5   | Brick                 | Compressive<br>strength linear<br>shrinkage and<br>water absorption                 | 5 minutes of<br>microwave sintering<br>with a power of 950<br>W at temperature of<br>900°C                        |
| [25]      | Red clay               | Drying               | 700  | 6, 8 and 10                                 | Brick                 | Density, cracks<br>generation and<br>water absorption                               | 8 minutes of<br>microwave drying<br>with a power of 700<br>W at temperature of<br>70°C                            |
| [26]      | Hwangtoh<br>clay       | Curing               | 40, 60 and 80  | 30, 60, 90,<br>120,150, 180,<br>210 and 240 | Paste                 | Internal<br>temperature<br>distribution,<br>porosity and<br>compressive<br>strength | 240 minutes of<br>microwave curing<br>with a power of 60<br>W   |
| [27]      | Kaolin clay            | Drying               | 100, 200, 300, 400,<br>500, 600, 700, 800,<br>900 and 1000 | -   | Brick                 | Deformation and<br>generation of<br>cracks  | 21 minutes of<br>microwave drying<br>with a power of 600<br>W for 3.5m, 200W                                      |

|  |  |  |  | for the next 6.5 m  |
|--|--|--|--|---------------------|
|  |  |  |  | and 100 W until the |
|  |  |  |  | drying completion   |

#### 766 2.4.1 Critical evaluation

The use of microwave heating for sintering, curing and drying of clay-based construction materials relies upon the uniform volumetric heating that directly penetrates the material leading to enhanced consolidation efficiency and accelerates densification, enhancing mechanical and durability performance [74-76]. This deep penetration combined with the uniform, volumetric heating can significantly reduce energy usage due to the short processing times and rapid heating rate [74-76].

The absorption of microwave energy depends to a large extent upon the chemical composition 773 of raw materials [26, 84]. It has been reported by Kim et al. [26] that the existence of high 774 content of SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> in raw materials will allow them to absorb microwave energy very 775 well and enhance curing of clay-based construction materials. Therefore, the total amount of 776 SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub> in the clay materials that have been cured or sintered with microwave were 777 more than 80% [24, 26, 83]. According to the reviewed studies, the chemical composition of 778 779 raw materials were not been reported for the studies that used microwave for drying of clay materials. 780

The reviewed studies in Table 3 show that the microwave processing time is reduced with increases in microwave power. Additionally, all the reviewed studies indicate a considerable reduction in processing time by using microwave heating compared to a conventional electrical oven. This is because the volumetric heating process is significantly more efficient in comparison with resistance heating [72, 85].

As heat is favourable for improving the performance of geopolymer materials, however, the use of conventional ovens is not an energy efficient technique as it takes a long time and consumes energy along with the negative environmental impact associated with it. Therefore, the produced geopolymer materials have used the microwave as an environmentally friendly

| 790 | source of heating. In the reviewed studies that employed the microwave as a curing technique,       |
|-----|---|
| 791 | the microwave powers varied significantly with 40, 60 and 80 W for [26] and 3000 W for [83].        |
| 792 | These variations in the power explain why the required processing time in [26] was 4 times          |
| 793 | the time required in [83].  |
| 794 | Regarding the utilisation of microwave as drying technique of brick, the reviewed studies have      |
| 795 | also showed a range of microwave powers that have been employed between 100-1000 W.                 |
| 796 | Additionally and similar to the use of microwave as curing technique, the utilisation of higher     |
| 797 | power resulted in reducing the processing time as the temperature inside the microwave is           |
| 798 | directly related with the microwave power.  |
| 799 | For the use of microwave as sintering technique, Taurino et al. [24] indicated that the utilisation |
| 800 | of microwave with a power of 950 W for 5 minutes was sufficient to produce brick with a             |
| 801 | compressive strength of 65 MPa.   |
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#### 818 **3. Discussion**

## 819 3.1 Characterisation of clay-based construction materials

Tables 1-3 exhibit the wide range of clay-based materials and methodologies that have beendeveloped and subjected to various research investigations.

A range of tests were carried out on the resulting products to evaluate their performance following different standards. Compressive strength was the common test considered by most of the reviewed studies as the compressive strength is considered a basic and universally acceptable unit of measurement to specify the quality of clay-based construction materials as stated by common standards [35].

Other tests were also used in assessing the performance of the products. For example, the water absorption test was conducted for most of the resulting bricks, blocks, stabilized earth blocks and compressed stabilized earth blocks as a durability measurement [24, 25, 35-37, 41, 42, 45-47, 64].

Since clay-based brick and blocks are recognised as materials which contribute towards the thermal insulation of buildings and consequently increase indoor comfort, some of the research projects [39, 43, 45, 51, 55] investigated this property. The results indicated that the thermal isolation of the clay-based unities have been reduced slightly with increasing the stabiliser content [39, 43, 45], however, the use of alkaline activators and especially in the presence of FA improved the thermal isolation of the clay-based unites [51, 55].

Freezing/thawing and wetting/drying tests were evaluated for different clay-based materials
produced in countries that experienced these conditions such as the United Kingdom, Spain
and Belgium. [23, 45-47, 62].

In order to identify potential toxic agents within the produced materials, some researchers conducted leaching test [53, 83]. Dassekpo et al. [53] have conducted leaching test to identify the Arsenic content as a toxic material because the raw materials were clay waste from construction sites. Additionally, Hájková [83] conducted the leaching for the alkali (K and Na) elements to decrease efflorescence of the mortar due to the alkali effect.

845 Different tests were conducted for evaluating the performance of the clay-based construction products for different application of microwaves. Porosity test was conducted when the 846 microwave was used as a curing technique because reducing the curing time by increasing the 847 temperature of curing can significantly produce larger pores with larger diameters and 848 consequently reduce the compressive strength at later ages [69, 83]. However, generation of 849 cracks were investigated when the microwave was used as drying technique due to the high 850 851 temperature of the core of the clay-based produces [25, 27]. Other tests such as linear shrinkage and internal temperature distribution were conducted when the microwave was used as 852 853 sintering and curing technique, respectively.

Although most of the reviewed studies aimed to reduce the negative environmental impact of the clay-based construction materials, only one research project actually investigated the environmental performance of the final product [45]. As such, more detailed environmental impact assessments of the other techniques will be required before they can be compared with conventional production methods.

# 859 3.2 Curing of clay-based construction materials

The term curing is a process that normally associated with the production of different construction materials such as concrete, mortar, stabilised earth blocks, etc. The curing process can significantly affect the performance of the construction materials and it usually aims at acquisition the full strength of developed product [86]. As the method of curing can be varied according to the process of production, therefore, this section will be divided into two sections:
curing of blended and stabilized clay-based construction materials and curing of geopolymer
clay-based construction materials.

# 867 3.2.1 Curing of blended and stabilized clay-based construction materials

Curing is very important process for stabilized clay-based construction materials, especially when cement or lime is used in preparation of these products as a stabilizing agent. Water curing would aid in enhancing the hydration process of the cement/lime as this process can take place more efficiently in the presence of water that results in hardening the materials treated with cement/lime. Table 1 shows the methods of curing that was used for each of the stabilized clay-based construction materials

According to the reviewed studies [36-39, 42, 44, 45], the produced clay-based construction 874 875 materials that incorporated cement and/or lime were kept moist for 28 days by immersing in water, spraying with water or placed in humidity chamber at ambient temperature that ranged 876 877 between 20-40°C according to the weather conditions in the country where the experiments were conducted. This is because the presence of moisture will allow unreacted cement or/and 878 lime particles to hydrate further, producing additional cementing gel as reported by Joel and 879 880 Edeh [86]. Additionally, increasing the age of cuing to 28 days or even longer [35, 46, 47] in the presence of moisture will aid the formation of secondary cementing gel that is formed due 881 882 to the chemical reaction between calcium from portlandite phase (CH) and silicates from the clays [2]. 883

# 884 3.2.2 Curing of geopolymer clay-based construction materials.

The method and period of curing can significantly affect the properties of the geopolymer claybased construction materials [17, 69]. Geopolymer clay-based construction materials are usually cured at ambient or slightly higher temperature after mixing. Normally, the curing temperature is preferable to be less than 100°C [17].Based on the reviewed studies [22, 23, 5164], different curing temperatures with various periods of times were employed in the
production of different geopolymer clay-based construction materials.

According to the reviewed studies, air curing was considered by many of the researchers under 891 ambient temperatures that ranged between 20-35°C. For the geopolymer clay-based 892 893 construction materials under air curing, the periods of curing was a minimum of 7 days and some of the studies extended the time to 28 days [55], 60 days [22, 60], 90 days [54, 63] and 894 895 even 180 days [58]. This is because the geopolymerisation reaction is very slow at ambient temperature and extending the period of curing is essential to produce materials with enhanced 896 897 strength and reduced water absorption due to the formation of additional geopolymerisation products [17, 22, 54, 58, 87]. 898

899 In addition, oven curing was one of the techniques used in the curing of geopolymer clay-based construction materials due to that fact that heat is essential to improve the reaction by 900 accelerating the dissolution of silica and alumina species from the raw materials [88-90]. 901 According to the reviewed studies, the curing temperatures were reported in the range between 902 40°C and 150°C and the period under oven curing ranged between 6 hours to 7 days. Slaty et 903 904 al. [62] reported that increasing the temperature (50°C, 60°C, 70°C and 80°C) improved the strength gain after 6-72 hours. However, high curing temperature (150°C) resulted in reduced 905 compressive strength due to the formation of large pores [64]. 906

907 In general, adequate curing for clay-based construction materials is required to produce
908 materials with good mechanical and durability performance to maintain their structural
909 integrity.

910

# 911 *3.3 Comparative assessment of the techniques used in the production of clay-based* 912 *construction materials:*

The sustainable development in the construction industry required evaluating different parameters of the production techniques. The criteria considered in this study for the assessment are environmental and economic aspects for the production of different clay-based construction materials. The study presents comprehensive evaluation of environmental impacts associated with different techniques to produce clay-based construction materials. Additionally, the study provide some facts about the economic feasibility of different production techniques.

## 920 3.3.1 Environmental assessment

From an environmental point of view, the construction industry is responsible for about 40% of the energy consumption worldwide, nearly 30% of the global greenhouse gas emissions, generation of solid waste, depletion of natural resources and environmental damage [91]. Therefore, the construction industry is looking for alternative techniques and materials with the aim of moving towards sustainable development. The main advantages of using sustainable techniques and materials are to protect the environment and ecology, reduce the depletion of natural resources, energy efficiency and healthy outdoor and indoor environment [91, 92].

The main goal of this study is to assess the environmental impact of different unfired claybased construction materials, and to compare them with traditional fired clay-based construction materials. The assessment criteria including quantifying the consumption of energy, consumption of natural resources, consumption of fossil fuel and production of greenhouse gases.

933 The production of clay-based construction materials through firing is considered the most934 significant impact on the environment. This is because of the high temperature kiln firing

needed that consumes significant amount of energy along with releasing large quantities of greenhouse gases including CO<sub>2</sub> (attributed to the utilisation of coal for the firing process)[5, 91]. Additionally, in some cases where the coal used for firing is of low quality, this could significantly contributes to the acidification due to the release of SO<sub>2</sub> emissions and the formation of NOx [91, 93]. Furthermore, the production of fired clay-based construction materials contribute considerably to the depletion of fossil fuels as firing obtained mainly from coal [91].

942 On the other hand, the production of unfired clay-based construction materials through stabilisation seem to be the trend to follow to achieve sustainable development in the 943 construction industry in terms of environmental concerns. However, the stabilisation technique 944 involves the addition of cementing material(s) such as lime or/and cement, whose manufacture 945 946 required intensive energy, consumes huge quantities of natural resources along with releasing huge quantities of CO<sub>2</sub> emissions [5, 91, 94]. The manufacture of Portland cement consumes 947 about 5.6 GJ of energy and requires approximately 1.5 tonnes of raw materials along with the 948 production of about 7% of CO<sub>2</sub> emission in the atmosphere [49, 95]. Additionally, the 949 production of stabilised clay-based construction materials contribute to larger water depletion 950 951 that needed for curing process and lower consumption of fossil fuels relative to fired clay-based 952 construction materials [91].

In comparison with the aforementioned production techniques, the production of clay-based construction materials through geopolymerisation consumes much less energy and releases considerably lower quantities of greenhouse gases [5]. Therefore, the environmental burden of the geopolymer clay-based construction materials are generally lower than the fired or stabilised clay-based construction materials [96]. However, the production of geopolymer claybased construction materials [96]. However, the production of geopolymer claybased construction materials is also associated with some environmental impacts that mainly attributed to the utilisation of alkali activators [97]. The manufacture of the alkali activators require intensive energy: (i) sodium hydroxide that is processed by electrolysis of salt water
and (ii) sodium silicate from the melting of soda ash and sand at about 1400°C [97].
Additionally, in order to achieve reasonable strength for geopolymer clay-based construction
materials, there is a need for curing at elevated temperatures (40°C-80°C) that means extra
energy consumption [5, 97].

## 965 3.3.2 Economic assessment

966 The feasibility of different production techniques should be evaluated in order to produce 967 sustainable products with competitive financial cost. The parameters included in the economic 968 assessment are the cost of raw materials and the energy required in the production.

The cost of raw materials can vary significantly among the production techniques. Poinot et al. 969 970 [96] reported that the cost of raw materials for fired clay-based construction materials could be 971 as small as 2% of the total cost of the final product, while in the case of geopolymer clay-based construction materials most of the cost is attributed to the alkaline activators. The highest cost 972 973 (about 60%) associated with the production of clay-based construction materials through firing is attributed to the consumption of large quantities of energy [96]. However, the unfired clay-974 based construction materials required smaller amount of energy to power the hydraulic pressure 975 976 compressed machines and for curing at elevated temperatures (for geopolymer clay-based construction materials)[5, 96]. 977

As overall, the evaluation of the environmental impacts and economic feasibility of different production techniques depends on considerations included in the assessment criteria and all of these considerations should be evaluated and compared to provide accurate evaluation of the performance of different clay-based construction materials. According to the evaluated criteria and from production point of view, the production of clay-based construction materials through geopolymerisation is seen to be the best production technique taking into consideration the low 984 environmental impacts and the potentially competitive financial cost relative to other985 production techniques.

### 986 3.4 Future trends

987 The sustainability index of different clay-based construction materials could be significantly 988 improved by employing cheaper and eco-friendly materials and manufacturing processes along 989 with concentrate on improving the mechanical and durability performance. The following are 990 some suggestions to improve the sustainability of different clay-based construction materials:

As the utilisation of cement and/or lime in the production of stabilised clay-based 991 construction materials reduced the sustainability index of the final products, therefore, 992 993 replacing these materials with viable alternatives, could significantly improve the sustainability performance of this manufacturing technique. The use of industrial, 994 agricultural and natural waste and/or by-products as partial or full replacement to 995 996 traditional binders (cement and/or lime) can lead to the production of stabilised claybased construction materials with superior environmental, financial and technical 997 benefits. Marcelino-Sadaba et al. [94] stated that replacing the cement and/or lime 998 partially by GGBS in the production of stabilised clay bricks resulted in reduced 999 environmental impacts and superior technical performance. 1000

Generally, the production of geopolymer clay-based construction materials have lower
 environmental impact, relatively similar cost to other production techniques and with
 relatively high mechanical and durability performance. However, the alkaline
 activators used in the production of geopolymer clay-based construction materials are
 considered as the main contributor to environmental impact and cost of this production
 technique. Therefore, the viability of this production technique could be further
 enhanced by; firstly, reduce the molar concentration of the alkaline activators, secondly,

1008 the utilisation of clay materials with a minimum Si/Al molar ratio of 2 to decrease the 1009 use of alkaline activator solutions and finally, utilisation of some waste materials with 1010 high alkaline content as alternative to traditional alkaline activators. Additionally, heat 1011 curing is essential for geopolymer clay-based construction materials to achieve 1012 adequate mechanical and durability performance, however, the use of conventional 1013 ovens is not an energy efficient technique as it takes a long time and consumes energy 1014 along with the negative environmental impact associated with it. Therefore, the use of microwave as an alternative source of heat can significantly reduce the cost and 1015 1016 environmental impact of this technology, as the microwave heating is uniform and 1017 volumetric, reducing energy consumption and curing temperatures, with very rapid heating rates and significantly reduced processing times, thus improving physical and 1018 1019 mechanical properties, and lowering environmental hazards.

# 1020 **4. Conclusion**

Based on the review of the research studies on the production of clay-based constructionmaterials, the following conclusions have been drawn:

A wide variety of clay types were investigated in the production of different clay-based
 construction materials.

The techniques studied for the production of clay-based construction materials were:
 blending and stabilising, alkali activation and the use of microwave heating as an
 innovative sintering, curing and drying technique. The method of blending and
 stabilising is based on replacing clay partially with some waste or by-product materials
 and adding cementing materials such as cement or lime. The technique of alkali
 activation is based on the chemical reaction between clay materials representing the
 alumina-silicate source and a high alkaline solution. The use of microwave heating is

based on volumetric heating that directly penetrates the material significantly reducingthe processing time.

- A detailed comparison between different production techniques were conducted in
   terms of environmental and economic aspects along with suggestion for future trend to
   improve the sustainability of different production techniques.
- In order to maximize the commercial production of clay-based construction products
   using the techniques discussed in this work, more research needs to be conducted on
   the environmental and economic benefits along with public education and
   standardisation.
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