

CHEMISTRY AS A KNOWLEDGE BASE FOR THE DEVELOPMENT OF SUSTAINABLE CEMENTITIOUS MATERIALS

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ABSTRACT

The globe is challenged with developing new materials that will guarantee resilience, sustainability, and performance of infrastructure. Of the materials globally in dire need of, to bridge the gross deficit in infrastructure, cementitious materials top the list. Their premier position is justified by the fact that they are required to build almost all forms of infrastructure needed to meet the sustainable development goals. Cementitious material like concrete has witnessed great evolution since it was first discovered and is still witnessing many innovations in its manufacturing processes. Despite this, cementitious material remains a major threat to climate change due to high greenhouse gases emission that is attributed to its production. Therefore, developing new materials that are environmentally friendly without compromising quality, is very important for the future development. In this article, overview of developmental stages of cementitious materials is presented, and the inevitability of cementitious materials for future development is equally established. The role of chemistry at every stage of development of cementitious materials is underscored. The paper further links the capacity to develop new materials to the understanding of chemistry of the materials. Similarly, capacity to deploy the knowledge of chemistry to this important area is also emphasized. It is concluded that chemistry is a sine qua non for future material development. [*African Journal of Chemical Education—AJCE 13(2), June 2023*]

CEMENTITIOUS MATERIALS IN HISTORY

Binder, or cementitious material in whatever form, has been the sine qua none of construction since existence of man on earth, which was put to be about two million years ago. Though at the initial stage of life, man used available materials to protect himself from harsh weather, while also migrating to regions of favourable weather [1].

In the early Neolithic period, man started putting materials together to build the first ever *permanent* structure. Since there was a need to provide wall and floor that will provide shelter, there in need for binding material to achieve a stone-like structure that can stand on its own. The loosely available materials then were sand, clay and other earthen materials, which must be bonded together to provide the needed structure. History has it that the plain mud, with or without straw, was the earlier binder that was just used, though the chemistry of its usage was not well understood then.

Some thousands of years after, which some authors put at about 9,000 years ago, inorganic binding materials were discovered. This discovery seemed to be coincidental and fortunately, it was found to be similar to the concrete found in the early structures built in the Galilee of Israel [2]. It was found that the “binder” was limestone-based, which hardened on adding water. Several other civilizations employed different material for binding loose earthen materials. While the Assyrians and Babylonians used raw clay, the Egyptians adopted lime and gypsum [1]. Furthermore, in another version of the use of binder in Egypt, it was reported that they used more of gypsum than lime

because there was lack of fuel to generate enough heat energy to decompose limestone into lime [3]. Egyptians utilized this material as binder for stone to build the magnificent pyramids around 2,600 years back. Materials that were mainly calcium carbonate containing silica, were often used in China as found at a location near Xi'an as far back as about 5,000 years ago [4].

Ancient Greek also used slaked lime with some volcanic ash as binder to pieces of rock around 1,000 BC. As for the Ancient Rome, based on the knowledge acquired from the Greek, they made monumental structures with binder made from burning of mixture of gypsum and limestone with plaster of Paris to produce hydraulic binder. Similar trend was found in some countries in Sub-Saharan Africa, where mud with straw was commonly used as building material. One thing that is certain from this trend is that mankind, based on experience, found that some materials if heated, could become hardened in the presence of water. Meanwhile, at these times, the knowledge of chemistry was too limited to offer explanation for the performance of these materials. Thus, the knowledge of chemistry was never the basis for the development, but experience. As time goes on, the chemistry knowledge based on experimentation with these materials advanced the understanding of the cementing performance of these materials, which subsequently reflected in the new binders that are being developed in the modern world.

Scientists began to understand the chemistry behind the cementitious materials used in the early age, by the earlier 17th Century. During this period, hydraulic cement surfaced. In about 1756,

after the Industrial Revolution, John Smeaton built Eddystone Lighthouse with stones bonded by mortar, which he produced from the burning of binary mixture slaked lime and clay [2]. In his experiment about 40 years after, James Parker burnt limestone-clay mixture up to a temperature of 1,100°C and ground the resulting product to produce a powder similar to the cement we have today. This is reported to be the foundation for the production of modern hydraulic cement.

Joseph Aspdin, who is known as father of Portland cement, refined the method of producing hydraulic cement. He mixed quicklime with clay in a certain proportion which he burnt to around 1200°C and added water to the mixture. Thereafter, he ground the mixture to fine particles, then dried it before reburning it in a shaft kiln [5]. This cement was later known as “Portland cement”. The name “Portland” was attributed to Portland stone found on the Isle of Portland, in South Dorset Coast. His invention serves as basis for rapid research in cement chemistry as a number of issues were generated from his method. Some of the questions that spur further research included the temperature at which the mixture should be burnt, and the proportion of quicklime and clay that should be used.

As of 1850, four cement plants were developed in UK, though production of Portland cement started in 1825. Portland cement production did not start in France until 1848, while it started in Germany and United States in 1850 and 1871 respectively [2]. In 1884, Isaac Charles Johnson conducted a separate study from which he found criteria to produce homogenous product, which can

be achieved by “burning the mix of limestone and clay at high temperatures of 1250°C or above until semi-molten” [5]. This was the foundation for the modern cement manufacturing. What is certain is that understating of cement chemistry is a complex one, thus improvement continues. The more the chemistry of the materials get clearer, the more the understanding and potential to improve on what has been earlier developed.

RELEVANCE OF CEMENTITIOUS MATERIALS TO INFRASTRUCTURAL DEVELOPMENT

As shown in the previous section, the use of cementitious materials started with the existence of man. As long as the man needs shelter, road and other infrastructure, there would always be a need for cementitious materials.

Rapid urbanization with geometric increase in population is a global challenge of the 21st century, posing a serious threat to livelihood. More than half of global population now live in the cities. By 2050, the number will increase by 75% with Sub-Saharan Africa having the lion share of rapid urbanization - with its global share rising from 11.3% in 2010 to 20.3% in 2050 [6].

According to the United Nations Population Division, the population density for Africa is projected to increase from 34 to 79 persons per square kilometer for the period between 2010 and 2050 [6]. The attendant import of this scenario is that the existing infrastructure in most urban cities

are strained beyond their carrying capacity, leaving a chunk of the population to vulnerability. Gross deficit in housing and gridlock on the highway as a result of limited paved roadway, as well as epileptic power and energy supply, are consequents of urbanization. All urban cities of the world are experiencing shortage of adequate housing, but more alarming in the rapidly urbanized cities in Africa. About 60 million housing units was found as deficit in Africa in the period of 2001 and 2011. Going by the increase in population, the gap between the demand and supply will subsequently increase. It is estimated that about \$63 trillion will be invested to meet the global infrastructure deficit [7].

A cursory look at the trend in the need for infrastructure indicates that substantial quantity of materials will be needed to build these structures. Materials like timber, steel, plastic, glass, bitumen as well as concrete (cementitious material) are very much essential. Nevertheless, of all these materials, concrete seems to be the only material that is environmentally friendly due to its relatively low carbon footprint compared to other materials (Figure 1).

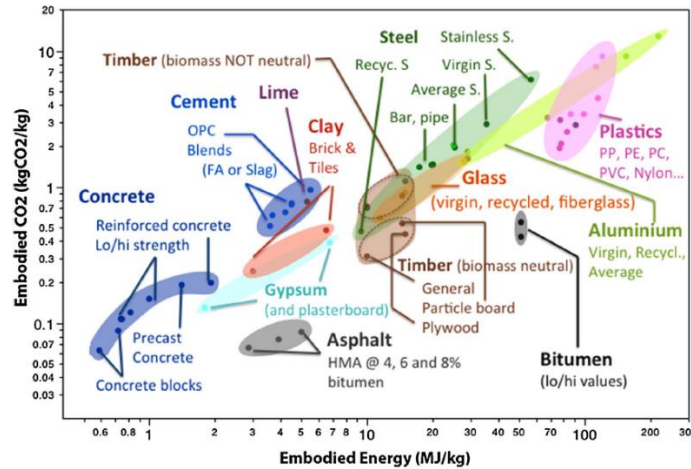


Figure 1: Carbon footprint of construction materials [8]

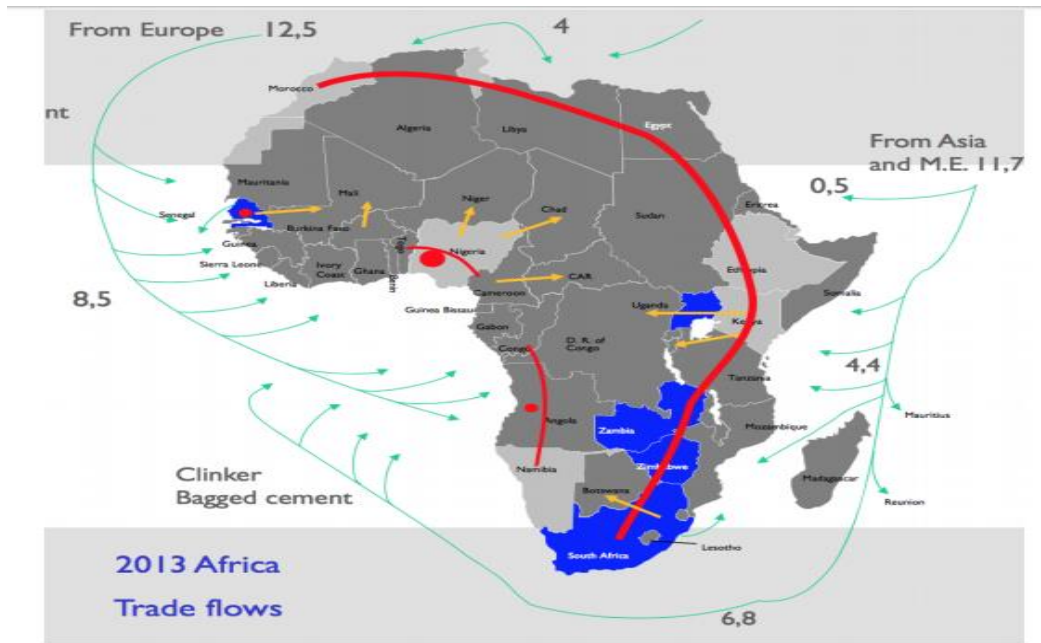
Concrete is a composite material that is needed much more than any other materials in construction industry. It is made up of cement, aggregates (fine and coarse) and water as well as other construction chemicals as the need may arise. Concrete has been in use for over 2000 years. The relative low cost, ease of production, possibility of forming different needed shapes and higher durability as well as low energy consumption gave concrete an edge over other materials (Table 1). In 2015, annual global cement consumption was 4.6Gt and this is projected to increase to between 6 and 13.5 Gt/a in 2050 [9]. Since concrete is needed to provide needed infrastructure, Africa has witnessed increase in investment in cement production. Figure 2 shows the network of cement distribution in Africa.

Though there is no alternative to cementitious materials in building the needed infrastructure, the huge consumption has created a big concern about the CO₂ emission from cement production.

Burning of fossil fuels and carbonation of limestone which is decomposed into lime and carbon dioxide are the major sources of CO₂ emission from Portland cement production. For a tonne of cement produced about 1 tonne of CO₂ is emitted [10]. This shows that to meet the estimated demand for cement of 50 billion tonnes in 2030 equal amount of CO₂ will be emitted into the atmosphere. Hence, there is urgent need to find alternative to cement, or create a better cement to ensure carbon net zero by 2040. No doubt, understanding the chemistry of earthen materials is a prerequisite to creating ecofriendly cementitious material.

Table 1: Estimated energy, water requirement and carbon emission between 2015 and 2030 [7].

Materials	Cumulative Material Demand (billion tonnes)	Energy	Water	CO ₂	Energy	Water	CO ₂
		Per tonne of material			In total		
		(kWh/t)	(Litres/t)	(kgCO ₂)	(kWh/t)	(Litres/t)	(kgCO ₂)
Cement	50.1	110	307	914	5,518	15,400	45,850
Steel	26.7	5,700	28,500	2,000	152,147	760,733	53,385
Aluminum	1.7	72,000	88,000	20,900	120,967	147,849	35,114
Total					278,632	923,982	134,349



Source: Lightart, 2014

Figure 2: Clinker distribution in Africa [11].

CHEMISTRY AS BASE KNOWLEDGE TO UNDERSTAND THE PERFORMANCE OF CEMENTITIOUS MATERIALS

Cement as well as concrete are man-made materials and as such, a knowledge of their chemical composition is very important for optimal performance in service. The chemical compositions of these artificial materials are so important that they are the basis for the production, classification, and selection of the materials for use.

The cement production process is an entirely chemical-based process which begins with the mining and crushing of limestone and other materials (calcium, silicon, aluminum, and iron oxides)

to produce a specific size and composition of the crushed powder. This raw material mixture is then pre-heated in cyclones to save energy and begin the dissociation of calcium carbonate (CaCO_3) into calcium oxide (CaO) before being sent to the kiln [12]. The heating temperature in the kiln is kept constant at around 1200-1450°C, during which calcium silicates and aluminates (Ca_2SiO_4 and CaAl_2O_4) are formed. This chemistry produces clinker, which is subsequently transferred to silos where it is pulverized and combined with gypsum to regulate the setting time of the cement produced [12 – 14]. The coagulating effect of gypsum can lead to poor dimensional stability and decreased strength when a high quantity (>5%) is mixed with the clinker in the kiln. On the other hand, when the quantity used is less than 3%, the retardation effect will be almost ineffective, thus making it necessary to specify an accepted range of around 3-5 wt.% of the cement composition.

Also, considering the effect of the milling temperature on the performance of the cement, it has been reported that the risk of producing a false setting cement can be reduced by any of the following: supplying the mill with a relatively cool clinker, recirculating cool air into the system or by using an internal water spray mechanism [15]. Considering the process enunciated above, one would realize that a sound knowledge of chemistry is essential to ensure that the performance of the product is as expected at all times. This is the same for other cementitious materials.

Portland cement is made up of four main crystalline components namely, Tricalcium silicate (Ca_3SiO_5), dicalcium silicate (Ca_2SiO_4), tricalcium aluminate ($\text{Ca}_3\text{Al}_2\text{O}_6$), and tetra calcium alumino

ferrite ($\text{Ca}_4\text{Al}_2\text{Fe}_2\text{O}_{10}$) [16] The concentration of these compounds in different cement types determines performance-oriented properties like strength, sulphate resistance, low heat of hydration, hydrophobicity, and resistance to seawater. When the focus is on strength development, it is standard practice to add various secondary elements, such as fly ash, silica fume, and granulated slag to the cement composition. If sulphate resistance is the priority, the tricalcium aluminate (C_3A) content is limited to a maximum of 8%. Whereas the addition of saturated fatty acids to the gypsum and clinker mixture produces a cement that repels water and is typically difficult to mix [17 – 19]. To achieve all these reactions, a careful and thorough understanding of cement chemistry is necessary. Table 2 summarizes all the chemistry of cement right from production up to the hardened stage of 28 days, when water is added. The table underscore the relevant of knowledge of chemistry in understanding performance of cementitious materials.

The understanding of cement chemistry is also essential during the hydration process. When mixed with water, each of these substances reacts to produce extremely potent hydration products, such as calcium hydroxide $\text{Ca}(\text{OH})_2(\text{s})$ and calcium silicate hydrate (C–S–H) as shown in equation 1-2. These hydrates form the basis for the selection of the most suitable supplementary cementitious materials to complement the basic compositions of ordinary Portland cement (OPC). While C-S-H produced from the hydration of cement aids cement's strength development process, $\text{Ca}(\text{OH})_2$ on the other hand is soluble and vulnerable to leaching. As a result of this, the pore structure of the

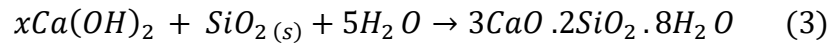
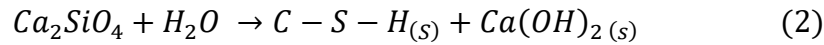
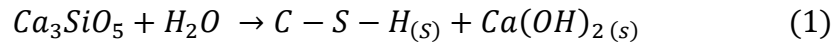
cement-based material is increased thereby weakening the cement matrix and reducing the compressive strength.

Table 2: Summary of chemistry of cement at different stages of production and use

Stage	Chemistry	Remarks
Calcination	$\text{CaCO}_3 \longrightarrow \text{CaO} + \text{CO}_2$	Major source of carbon emission
Clinkering	$2\text{CaO} + \text{SiO}_2 \longrightarrow \text{Ca}_2\text{SiO}_4$	Dicalcium silicate (C_2S) is formed and represent 45 – 75% of mass of clinker
	$3\text{CaO} + \text{SiO}_2 \longrightarrow \text{Ca}_3\text{SiO}_5$	Tricalcium silicate (C_3S) is formed and represent 7 – 32% of mass of clinker
	$3\text{CaO} + \text{Al}_2\text{O}_3 \longrightarrow \text{Ca}_3\text{Al}_2\text{O}_6$	Tricalcium aluminate (C_3A) is formed and represent 0 – 13% of mass of clinker
	$4\text{CaO} + \text{Al}_2\text{O}_3 + \text{Fe}_2\text{O}_3 \longrightarrow \text{Ca}_4\text{Al}_2\text{Fe}_2\text{O}_{10}$	(Tricalcium aluminoferrite (C_4AF) is formed and represent 0 – 18% of mass of clinker
1 st Day Hydration	$\text{C}_3\text{A} + 6\text{H}_2\text{O} \longrightarrow \text{C}_3\text{A}.6\text{H}_2\text{O}$	It is an exothermic reaction with release of about + 880 kJ/kg energy.
Early Setting	$\text{C}_3\text{A} + 3\text{CaSO}_4.2\text{H}_2\text{O} \longrightarrow \text{C}_3\text{A}.3\text{CaSO}_4.2\text{H}_2\text{O}$	To avoid early setting, gypsum is added
2 – 7 Days Hydration	$2\text{C}_3\text{S} + 6\text{H}_2\text{O} \longrightarrow \text{C}_3\text{S}_2.3\text{H}_2\text{O} + 3\text{Ca}(\text{OH})_2$	Tobermonite gel is formed, which has high surface area and high cementing property with release of energy of about + 500 kJ/kg
7 – 28 days of Hydration	$2\text{C}_2\text{S} + 4\text{H}_2\text{O} \longrightarrow \text{C}_2\text{S}_2.3\text{H}_2\text{O} + \text{Ca}(\text{OH})_2$	Hardened material is formed with about 90% of strength achieved and released of + 250 kJ/kg energy

Furthermore, the weakened matrix allows the ingress of hazardous ions (SO_4^{2-} and Cl^-) into the concrete, which initiates spalling and steel corrosion in reinforced concrete [20]. For this reason, the selection of silica (SiO_2) rich supplementary cementitious materials as a measure to reducing the quantity of hydrated $\text{Ca}(\text{OH})_2$ and producing more C-S-H in the concrete mix, is a common practice

among concrete experts to improve the durability, resistance to acidic and chloride attacks. The summarized chemistry of the reactions is shown in equation 3. Therefore, it is convenient to say that understanding the basic chemistry of cement and supplementary cementitious materials (SCMs) is very essential to fully grasp the relationship between the material's composition and performance.



MODERN CEMENT AND CLIMATE CHANGE

Recent studies have shown that the global cement sector is a significant source of industrial greenhouse gas emissions, accounting for around 5-7% of all anthropogenic global warming emissions [21 - 22]. With the current spate of emissions, it is believed that CO₂ emissions must be reduced by half before 2050, if the sustainable development goals on climate change must be achieved. This reverie is rather amusing, given that worldwide CO₂ emissions from cement plants have tripled in the last two decades and are still rising as at the time of writing, as shown in Figure 3; with countries like China, India and United States of America topping the list of main contributors to annual CO₂ emissions from cement industries.

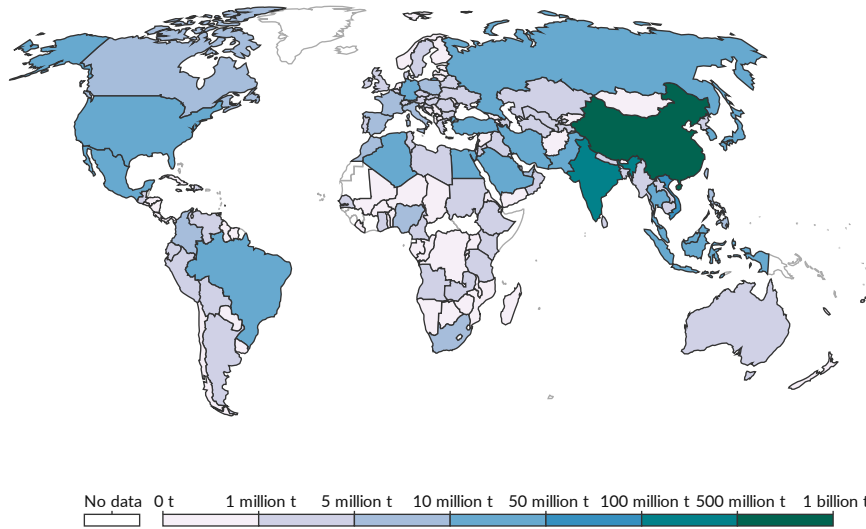


Figure 3: Annual emission of CO₂ by countries from 1880-2020 [23]

Given this trend and the exponential rate of the modern industrial revolution, CO₂ emissions from the global cement sector will be around 25 billion tons by 2050. What is more worrisome however, are some of the damages that this phenomenon will inflict on nature if not addressed [24 - 25]. Therefore, it becomes imperative for the modern cement industry to begin to adopt technologies and methods that promote carbon neutrality from the production of cement to its final drop in concrete elements. One recent discovery that has a strong potential to promote the decarbonization of cement is the use of biogenically manufactured limestone as a replacement for conventional limestone. This material is biologically produced from microalgae through photosynthesis [26].

Findings from a recent study have shown that if the current global demand for limestone could be met by biogenically generated limestone, the annual CO₂ emissions from cement and concrete manufacturing would be reduced by 2 gigatons.

The use of Graphene-based Nano Sheets (GNS) reinforcement has also been reported to be an effective method of addressing the environmental difficulties associated with cement and concrete production. According to Basquioto de Souza et al [27], this is conceivable because of GNS's superior mechanical, permeability, and densification capabilities in the cement matrix. By using as little as 0.05% of GNS, concrete's mechanical and durability properties can be improved by as much as 80% and 500%, respectively, thereby resulting in a reduction in the demand for OPC in ordinary concrete. Furthermore, its exceptional electrical and thermal properties make it an excellent choice for use as a smart property inducing agent in concrete [28 - 29]. Considering its impressive performance, it is reported that if GNS could replace OPC by up to 50% while maintaining building loadings, this would result in a 0.45-tonne CO₂ reduction for every tonne of cement produced [27].

Another alternative with great potential for CO₂ reduction is the use of alkali-activated geopolymers as binders in concrete. These binders can outperform typical cementitious binders in a range of applications while producing significantly lesser amount of greenhouse gas. Furthermore, alkali-activated binders have been recognized for their excellent resistance to high temperatures and

high thermal insulation, shorter curing period and increased durability in harsh environments [30 – 32].

NEW MATERIAL AND DEPLOYMENT OF KNOWLEDGE OF CHEMISTRY

It is evident from the discussion above that a thorough understanding of the production process as well as the chemical properties of cement is crucial to developing a sustainable solution to the carbon emission issue. As reported by Czigler et al. [33], improvement in operational advances can only reduce emissions by approximately 20% from their current levels. Therefore, if there is to be a considerable reduction in CO₂ from cement related activities by 2050, new line of technologies and alternative materials must be developed in order to achieve that goal. In addition to this, multidisciplinary approach among diverse specialists in the cement sector would also be beneficial as this will help to have a broader view of how to address the decarbonatization of cement across various disciplines. Furthermore, since a significant portion of the CO₂ produced at cement plants comes from the calcination of clinker, efforts should be made to develop smart cement factories by implementing Carbon Capture and Sequestration (CCS) systems on-site. As such, instead of releasing the potentially dangerous greenhouse gases into the atmosphere, the plants can self-capture the CO₂ and store it for use as biofuels, carbamate derivatives, carbonates, polycarbonates, and carboxylic acids, which are raw materials used in power generation, the paint industry, and

pharmaceuticals [34 - 35] The clinker can also be substituted with other materials that have similar properties and lower carbon footprints.

On the part of the material, the successes recorded in the adoption of carbon-neutral materials like biogenically produced limestone, limestone calcined clay cement (LC3), alkali-activated geopolymers and rapidly evolving materials like graphene should be built on and scaled up for larger-scale implementation. Also, as shown in Figure 4 emphasis should be placed on the use of alternative building materials such as cross laminated timber (CLT) which has been reported to have high mechanical and temperature resistance and is capable of reducing the carbon footprint by nearly 25%, by simply using 10% of CLT as against cement-based structure [33].

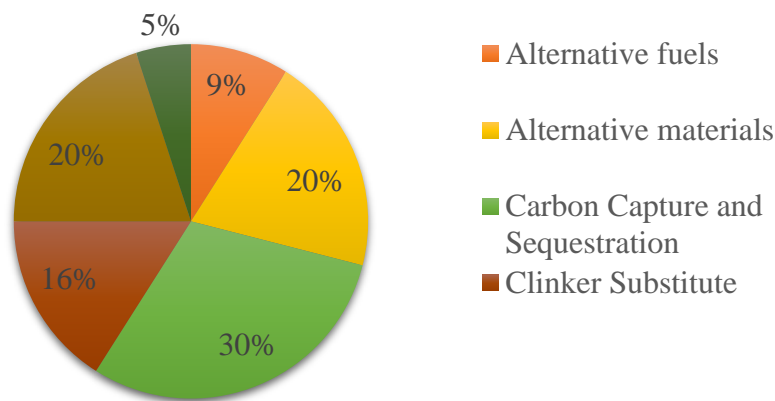


Figure 4: Projected reduction in CO₂ from cement industry as a result of innovative technologies by 2050 [35]

CONCLUSION

Cementitious material is an age-long material known in the history of man. The material is indispensable as there is need for binding loose earthen materials for building structures. Almost all the infrastructure needed for good livelihood needs cementitious material as one of its ingredients. Though the usage of the material in the early age was based on mere practice and experience as against understanding the science of performance. As the knowledge of chemistry of materials grew the principle behind the formation of artificial stone became clearer. Thereafter, scientist deployed the knowledge of chemistry to develop new cementitious materials that are less costly to the environment. Hence, knowledge of chemistry played and will continue to play crucial roles in furthering the understanding of cementitious materials.

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