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Impact of incorporation of volcanic ash on geopolymerization of eco-friendly geopolymer composites: A review

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Abstract. Volcanic ash is a product from an explosive type of volcanic eruptions. Fresh particles of volcanic ash are gritty, abrasive, vexatious and corrosive with huge scale dispersion. Consequently, it is the need of the hour to dispose this waste systematically to have relief from dilemmas like land fillings; climatic changes, pollution of the environment, water, and health hazards although the soil is a mineral intake. This crucially reviewed manuscript includes not merely the comprehension of the incorporation of volcanic ash to develop novel green Geopolymer composites but also to study its impact on the geopolymerization reaction kinetics and reactivity at dissimilar temperatures along with a precise account of its chemistry, mineralogy and the morphology.

1. Introduction

Currently, infrastructure and construction industries are experiencing a nimble escalation due to the burgeoning population of the world. Consequently, the exigency for concrete as a sine qua non-construction material has also intensified drastically [1-3] owing to its exponential application that, in turn, boosted the demand for Ordinary Portland Cement (OPC) as the dominant binder of concrete. Disastrously, the current practice of OPC production is not merely immensely energy intensive and necessitates elevated temperature which is obtained using natural coal fiery but also emits deleterious gases like CO₂, SO₃, Nitrogen oxides, etc., into the atmosphere, consequential in the acid rain at times and global warming [4-7]. For illustration, the production of a ton of OPC liberates an approximately equal amount of CO₂- a principal greenhouse gas, into the environment and devours about 4GJ of energy [7-11]. Merely cement industry alone is accountable for roughly 7% of total CO₂ emissions across the planet [7,12,13]. This is a solemn peril to the plants, animals and humanity across the sphere. Concentrated research endeavours are being dedicated to the invention of eco-friendly construction materials.

Geopolymers are novel inorganic class of materials produced by a geopolymerization process of polycondensation upon reaction between an alumina silicate (rich in silica and alumina contents) and



an alkaline solution at low temperature. The reaction consists of dissolution, speciation equilibrium, gelation, restructuring and polymerization – a process analogous to rock-forming geosynthesis [4-6, 14], to produce reticular molecular structures comprised of four coordinated Si and Al units [15-19]. Geopolymer concrete exhibits marvellous physical, optical, mechanical and thermal characteristics; geopolymer formation involves very low energy consumption (up to 60 percent less than OPC) [20-22, 23-27], thus geopolymers form as an eye-catching substitute to conventional OPC-concrete. The attributes embrace not merely exceptional stability at an elevated temperature, tiny shrinkage, freeze/thaw, autoclave curing, resistance to acid and fire, etc.; but are also significantly often cost-effective (10%-30%) and with low carbon footprints (nine times lower) as compared to conventional concrete [5,14,27-31]. A range of Geopolymer concrete can be developed with diverse resources of materials, but the best-desired sources are varieties of wastes either artificial or natural like farming, geological waste products, etc. Quite recently, one of the innovative perceptions is of systematic application of volcanic ashes as source material to develop a novel Geopolymer concrete.

When a volcano erupts, fragments of pulverized volcanic minerals, rocks, glasses, etc. possessing grain size less than 2 mm, (i.e., 0.2 cm., or 0.079 inches,) in diameter jetted violently into the atmosphere, are known in common term as “Volcanic Ash”. Therefore, they are the product of explosive volcanic eruptions. The dissolved gases present in magma expand resulting in an aggressive flee when reaching the surface of the earth. Also, they can be produced through a phreatomagmatic eruption which is an explosive interaction of water bodies like the sea, lakes, marshes, groundwater, snow, ice, etc. and magma whereby huge quantities of magmatic gases and steam are liberated vigorously resulting in shattering of magma. They may also be formed all through transport in pyroclastic density currents, i.e., a swiftly-moving flow of hot gases along with volcanic matters which travels away from the volcano. In this case, particle abrasion observes as the particles interact mutually resulting in a lessening of grain size as well as the creation of fine-grained volcanic ashes.

Fragmentation causes a boost in the contact area between water and magma which results in a feedback mechanism,[32] directing to supplementary fragmentation and production of fine volcanic ashes. Volcanic ashes can be thrown several kilometres from the vent of the erupting volcano which may subsequently transport by the wind for thousands of kilometres away. They are a significant volcanic hazard and can cause severe predicaments owing to their extensive dispersion. Mushrooming growth of human population is accountable for the escalating development of urban areas into high-risk zones such as surrounding areas of volcanic centres that mostly results in volcanic ash fall tragedies. They also can generate a cloud of ashes and turn the fresh air into irrespirable to lives of the earth accounting for asthma, chronic lung diseases, other health hazards to human, animals, etc.

Additionally, these ashes may found as huge eruptions preserved and can have a lot of pessimistic impacts on humans and animals health, electricity, large and small water treatment systems, wastewater treatment plants, supplies of rainwater-fed and drinking water, interruptions in functioning of aircraft and airport systems, computers, land transports, buildings and structures, telecommunications, environments and agriculture etc. The sheer weight of deposits of falling ash is responsible for serious damage to vehicles and infrastructures. Therefore, the systematic consumption of these gigantic quantities of volcanic ashes would be advantageous to reduce the environmental crisis. The construction and infrastructure industries can swallow these volcanic ashes in high volume by employing them as pozzolans to produce blended cement or as lightweight ingredient materials to manufacture Geopolymer concrete and mortar, i.e. Volcanic ash - based Geopolymer composites.

Surprisingly, there is scanty data accessible on the topic of applications of volcanic ashes in manufacturing Geopolymer composites which may be due to its existence in only restricted countries of the world! No doubt there found attempts to employ it as supplementary material in conventional composites, but in the case of Geopolymer composites, it is employed as chief ingredient resulting in a possibility of its gigantic consumption.

This manuscript not merely reviews the existing literature on volcanic ash-based geopolymer composites and facilitates to understand better sustainability and appropriateness for geopolymeric

synthesis along with a critical evaluation of existing research works highlighting the obstacles in the pathway for its application but will also direct for future research work on the subject matter.

2. Physical and chemical characteristics of volcanic ash

The physical and chemical attributes of volcanic ashes depend primarily upon the nature of the volcanic eruption, chemistry, crystal content, embodied gasses, temperature etc. of the lava ejecting from a variety of fashions of eruption exhibited by volcanoes [33].

This can be classified using the Volcanic Explosivity Index (VEI). Violently explosive eruptions (VEI 5+) of acidic nature and Rhyolitic, as well as Dacitic composition, can bring about massive amounts of the ejecta of more than 10^9 m³ while Effusive eruptions (VEI 1) of basic nature Basaltic composition produce less than 10^5 m³ of ejecta into the environments. Another parameter to manage the quantity of these generated ashes is the period of the eruption, i.e. the longer sustainability of the eruption will produce more volcanic ashes. For instance, in 2010, the 2nd phase of eruptions of the volcano of Eyjafjallajökull was considered as VEI 4 even though a modest eruption column of 8 km height were recorded. This happened to owe to the prolonged period of the eruption which continued for a month permitting a huge volume of ashes to be ejected into the environments.

The chemical properties of volcanic ashes are reliant upon the kinds of accompanying minerals and the chemical composition of the erupting magma. The Siliceous magma contains Silicon and Oxygen profoundly, and consequently, volcanic ashes generated through volcanic eruptions of this type of magma are generally described in terms of their content of Silica. Whereas, the eruptions of Basaltic magma with low energy are yielding archetypal volcanic ashes with a dark colour having 45 - 55% Silica, mostly rich in Iron and Magnesium contents. On the other hand, the most explosive acidic natured Rhyolite eruptions generate felsic volcanic ash with a more higher Silica content of more than 69% and some other intermediately composed volcanic ashes like Andesitic or Dacitic have Silica content somewhat ranging between 55-69%.

The Water vapours, CO, CO₂, SO₂, H₂, H₂S, Hydrogen Chloride, etc. are the primary gases emitted for the duration of volcanic activities [34]. Moreover, Sulphur and Halogen gases and metals are being taken away from the environment by the chemical reaction processes, wet and dry deposition, as well as by adsorption on the exterior of volcanic ashes.

A variety of Sulphates and Halides, i.e. principally Fluorides and Chlorides compounds, are enthusiastically rallied from fresh volcanic ashes which is a long-recognised fact [35-37]. These salts are believed to be formed owing to the quick dissolution of ash particles in acid within eruption plumes, which is also supposed to provide the cat-ions engrossed in the deposition of salts of Halides and Sulphates.

The leachates from fresh volcanic ashes deposits may embrace nearly 55 ionic elements [34] prominently like the cations of K⁺, Na⁺, Mg²⁺, Ca²⁺ etc. and the anions such as SO₄, F⁻, Cl⁻, etc. [34,37]. In many instances these elements represent as simple salts viz., CaSO₄, NaCl etc. as suggested by the molar ratios between ions found associated with these leachates [34,38-40]. The eruption of Mount St. Helens in 1980, Chloride salts were met with as the most enthusiastically soluble subsequent to sulphate salts in an experiment carried out on fresh volcanic ashes in sequential leaching [38]. Whereas, Fluoride compounds like CaF₂, MgF₂ etc., generally, found as sparingly soluble except Fluoride salts of alkali metals and compounds such as CaSiF₆ (Calcium Hexafluorosilicate)[41]. The pH of the fresh volcanic ashes leachates may vary extensively as to the amount present of acidic gas condensate on the surface of the ashes (principally as a result of the SO₂, HF and HCl gases in the eruption plume. XRD image of volcanic ash is represent in Figure 1.

The role of the crystalline-solid structure of salts was found more like an insulator rather than a conductor [42-45]. Nevertheless, the volcanic ashes may turn to be corrosive and electrically conductive, once the salts are dissolved into a solution through a resource of moisture such as light rain, fog, mist, etc. The electric conductivity of volcanic ashes enhances with:

- (1) Boosting the content of moisture,
- (2) Augment insoluble salt content, and

(3) Advancement in compaction (bulk density) as investigated in a recent study [45].

The close up view of a small particle of volcanic ash, displaying its many small tubular holes. The particle of volcanic ashes erupted during magmatic eruptions from Mount St. Helens are comprised of fractions of vitric, i.e., glassy and non-crystalline, crystalline or lithic, i.e., non-magmatic particles. The volcanic ashes produced during magmatic eruptions of low viscosity produce a variety of diverse pyroclasts depends upon the process of eruption. A considerably more viscous eruption like Basaltic one forms a range of pyroclasts from a symmetrical droplet of sideromelane to blocky Tachylitic type. In contrary, most volcanic ashes with high-silica content comprise of pulverised Pumice yielding sand a few lithic fragments along with individual phenocrysts [46].

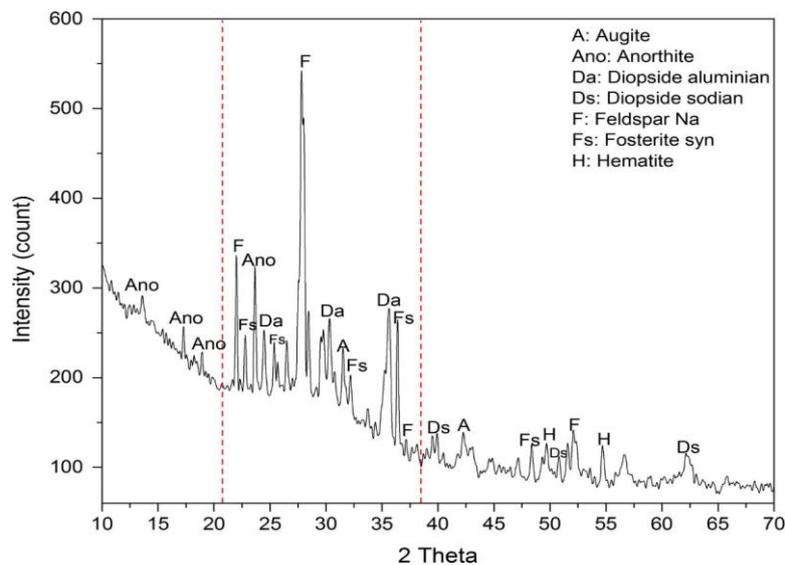


Figure 1. XRD of volcanic ash [49].

The volcanic ashes are mainly found comprises of mineral fragments and hydro-thermally altered lithic mostly in a matrix of Clay when their production occurs during phreatic eruptions. Here, the surfaces of the particles are frequently found coated with Clay or ingredients of Zeolite crystals and the identification of types of Pyroclast is done with only relict textures [46].

3. Morphology of volcanic ashes

The morphology meaning the shape of volcanic ashes is depended upon a plethora of diverse eruptions and kinematic processes [46,47].

Eruptions of less viscous basaltic magmas form droplet-like particles of volcanic ashes which are partly controlled through surface tension, the friction of air, an acceleration of the droplets after leaving the vent of a volcano. The shapes are found ranging from a perfect sphere to variously twisted, elongated droplets having even fluidal surfaces [47].

While the morphology of volcanic ashes from eruptions of highly viscous magmas dependent mostly upon the shape of vesicles in the upcoming magma prior to disintegration. The vesicles are produced through the spreading out of gases embodied with magma prior to solidification of the magma. The particles of volcanic ashes can have dissimilar vesicular particles and vesicularity and also enormously high ratios of surface area to volume [46].

The broken vesicle walls are formed owing to concavities, tubes, and troughs observed on surfaces of grain [47]. Angular, vesicular fragments that are pumiceous or thin vesicle-wall fragments are found occurring in vitric volcanic ashes generated from the eruptions of highly viscous magma. Whereas, the lithic fragments of volcanic ash are found angular to sub rounded in shape. Lithic morphology in volcanic ashes is commonly depended upon the mechanical properties of the wall rock

which are shattered by the explosive expansion of gases embodied in magma or spalling when it touches the surface.

The morphology of volcanic ash particles generated through phreatomagmatic types of eruptions is managed by forces present in the chilled magma that result in divisions of the glass to yield small pyramidal or blocky particles of glass ash [48].

In the case of phreatomagmatic types of eruptions, merely a trivial role in the determination of grain shape is played by the density and vesicle shape. In this case, the coming up magma is cooled swiftly when coming into contact with surface or groundwater. The principal kinds of pyroclastic shapes of fragments formed due to the forces in the "quenched" magma are (1) vesicular and irregular but having even surfaces; (2) equant and blocky (3) drop-like or sphere-shaped (3) convoluted and moss-like and, (5) plate-like.

4. Impacts of chemistry and mineralogy of volcanic ashes on geopolymerization

4.1. Chemistry of volcanic ashes

Looking to the chemistry of volcanic ashes, they are found to be made up of small petite fragments of jagged rock usually in association with diverse minerals, and volcanic glasses. They are hard, abrasive and insoluble in water. Table 1 demonstrates the chemical composition of a few volcanic ashes employed for geopolymer synthesis based on obtained from previously published papers.

The greater Loss on Ignition (LOI) monitored in a few volcanic ashes indicates the quantity of dissolved gases like CO₂ and H₂O in the glassy phase in a dissimilar form [36]. The presence of H₂O points towards the content of clayey minerals, whereas CO₂ may be in carbonate form or entrapped in the glassy phase as CO₂ molecules [49,50].

4.2. Mineralogy of volcanic ashes

Glancing at the mineralogy of volcanic ashes, the kinds of minerals found accompanied with them are reliant on the chemistry of the erupted magma.

Table 1. Typical chemical composition of volcanic ashes (Formatting sections, subsections and subsubsections).

Chemical Composition	Andesite	Volcanic Scoria	Volcanic Pumice
CaO	2.52-6.69	9.07-11.00	0.87-6.79
SiO ₂	54.93-61.57	40.17-46.48	64.67-70.13
Fe ₂ O ₃	4.93-10.89	12.00-15.16	1.27-3.03
Al ₂ O ₃	16.44-19.00	13.86-15.41	11.11-14.06
Na ₂ O	1.65-6.00	3.39-4.6	0.11-2.30
K ₂ O	1.72-1.95	0.9-1.53	2.25-4.26
MgO	2.63-4.25	6.45-9.57	0.25-1.11
LOI	2.15	0.66-9.31	3.94-10.28

Generally, its mineral contents can be classified as non-magmatic and magmatic [51]. Magmatic minerals are originated from Silicate melt and crystallize during the eruption activities while non-magmatic minerals incorporate unintentionally with volcanic expel owing to the interaction between crustal rocks and magma. As a result, any minerals forming crustal rocks may be found associated with volcanic ashes [48,51,52]. Non-magmatic minerals are formed from weathering processes of volcanic glasses include Mica and Clay Minerals.

4.3. Geopolymerization and the Reactivity of Volcanic Ashes

Reactivity of aluminosilicate source material for Geopolymerization synthesis is its capability to dissolve with ease in an alkaline medium which depends primarily on the fineness of particles and

mineralogical as well as chemical compositions of the aluminosilicate. In the study of Lemougna et al. [53], the effect of mineralogical and chemical compositions on the reactivity of four volcanic ashes from Cameron utilizing differential scanning calorimetry. The heat liberated in the course of the reaction is dependent on the mineralogical composition of volcanic ashes.

Tchakouté et al. [54] noted the relationship amongst the structural water possessed in two volcanic scoriae, Clayey mineral and the LOI as well as their reactivity. In conclusion, they revealed that the presence of Muscovite in one of that volcanic scoria and structural water was responsible for its comparatively enhanced reactivity.

Djobo et al. [55] have also investigated the reactivity of volcanic ashes by confirming the composition of the glassy phase and the quantity along with the entire heat released employing ICP-OES and Iso-thermal Conduction Calorimetry (ICC) correspondingly. They noted a very inferior dissolution of it in alkaline solution and heat release met with was low throughout geopolymerization maintaining the same conditions. They concluded that it was due to its inferior reactivity. For that reason, it is palpable that some of the characteristics of LOI and Clayey minerals associated with volcanic ashes are responsible for controlling its reactivity. On account of the inferior reactivity of volcanic ashes, many researchers suggested the procedures to modify it.

The Clayey mineralogical composition of volcanic ash can only favour Calcination. Bondar et al. [56] accounted that a suitable an apposite Calcination temperature is essential to obtain modifications in volcanic ash, that depends upon the reactivity and nature of Clayey minerals content. Consequently, a unique Calcination temperature that might be appropriate for changing the reactivity of all volcanic ashes was not set up. The process of Calcination is energy intensive since it needs a temperature ranging between 700°C and 900°C.

While for the process of alkali fusion, Tchakouté et al. [57], suggested relatively low energy consumption requirement as the optimum Calcination temperature is found to be 550°C. But the surplus NaOH (Sodium Hydroxide), had not taken part in the reaction and necessitated being devoured by adding-on another aluminosilicate like Metakaolin. Dissimilar to both alkali fusion and Calcination, mechanical activation enhances not merely the quantity of amorphous phase but also so many reactive centers, that are in a greater energy state than in the raw volcanic ashes.

Furthermore, Calcination predominantly affects the clayey minerals while during mechanical activation, all minerals are exposed to structural alterations [58].

Thus, Calcination is less effective than mechanical activation. A few researchers have reported that the study of the kinetics of geopolymerization of volcanic ashes through measurement of the heat discharged during the reaction.

The kinetics of Geopolymerization of volcanic ashes at 25 °C with Ground Granulated Blast-Furnace Slag (GGBS) as additive employing Isothermal Calorimetry, and examined that the reaction is swifter with adding up of GGBS slag as investigated by Robayo et al. [59].

Similarly, Djobo et al. [49] noted the kinetics of Geopolymerization of volcanic ashes for temperature ranging from 27 to 60 °C employing ICC (Figure 4).

As to them, the greatest rate of reaction was achieved within 30 minutes at 27 °C and was delayed when the temperature goes up to 60 °C. Quite recently, DSC was employed for estimating the kinetics of Geopolymerization of volcanic ashes at 65°C, 75°C, and 85 °C [60].

Lemougna et al. [61] demonstrated the Calcium distribution which is a homogenous one and little spreading of Magnesium all through the mixture of volcanic ash based Geopolymer (Figure 2).

Other authors [62-65] have investigated the composition of gel of Oyster shell cum volcanic ash based alkali-activated system at various conditions of synthesis by CaO, SiO₂, and Al₂O₃ ternary diagram (Figure 3).

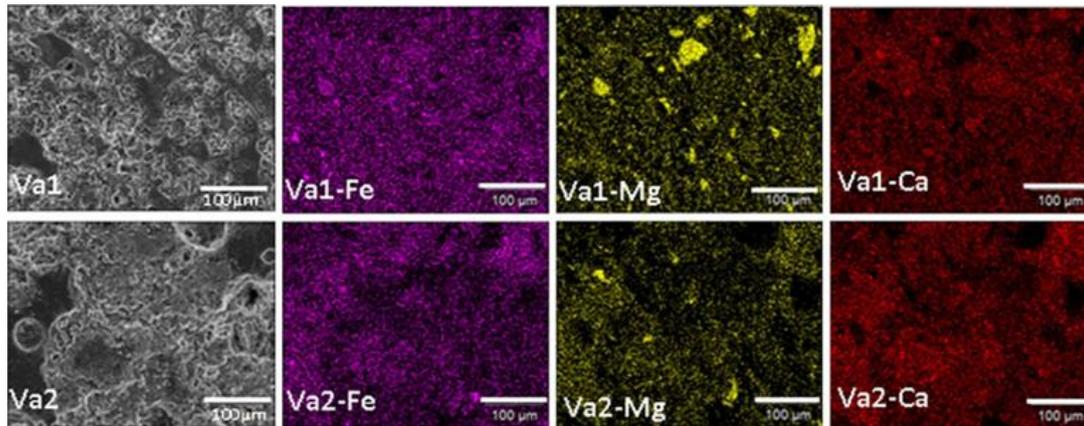


Figure 2. SEM image and EDS of alkali-activated ashes [61].

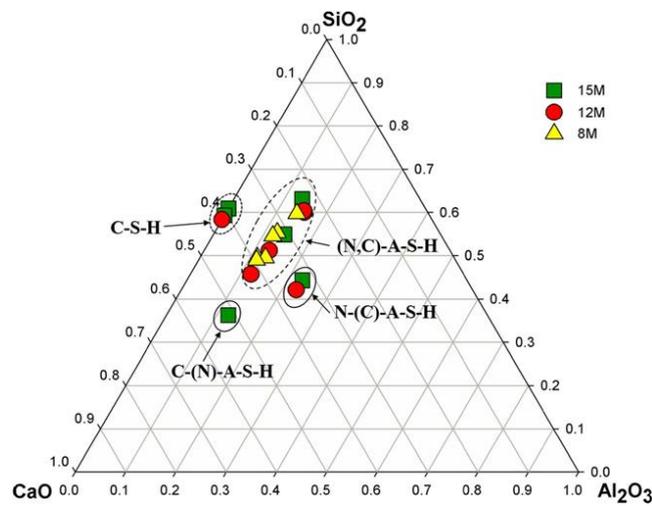


Figure 3. Illustration of EDS analysis of alkali-activated oyster shell volcanic ash [62].

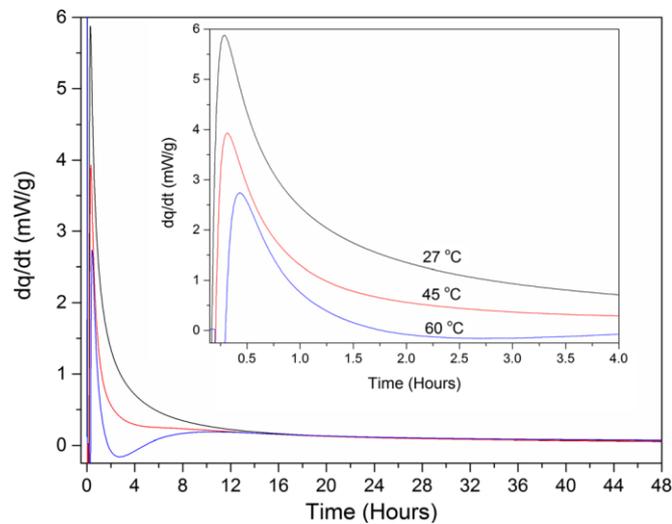


Figure 4. Reactivity of Volcanic Ashes at different temperatures [48].

5. Conclusion and perspectives

Volcanic ashes can be considered as appropriate and sustainable source material for developing novel Geopolymer composites viz., Volcanic ash based Geopolymer concrete, Geopolymer mortar and Geopolymer cement through Geopolymerization process. Its application proved to be favorable owing to its easy and profound accessibility in some parts of the world and lowering down pessimistic impacts on environments along with cost-effectiveness. Nevertheless, the present review has represented that these source materials are inferiorly employed as compared to other available aluminosilicates on account of the priority given by so many researchers forgetting rid of the predicaments of landfilling and disposal of some gigantic amount of wastes from industries like Slag, Fly ash etc. Also, there prevail some misapprehensions regarding the topic of temperature reliance of Geopolymerization of volcanic ashes which are needed to be examined methodically yet. Volcanic ash-based geopolymer composites viz., GP-concretes or GP-mortar or GP-cements, exhibited remarkable mechanical and physical attributes as well as brilliant durability as compared to OPC. Even though, research works reviewed in this manuscript demonstrated their appropriateness as construction materials, some supplementary researches are still called for to be executed to fill up the hiatus of data and swell the knowledge for its promotion analogous to Fly ash, Slag, Metakaolin, etc. based Geopolymer composites. The stress should be put on the topics of durability testing viz., Carbonation, alkali-silica reaction (ASR), and Freeze-thaw resistance. Also, volcanic ash based geopolymer composites necessitate being investigated in advance with fibre reinforced Geopolymer to evaluating their appropriateness for application for structures. So far, Geopolymer composites developed with this lightweight and porous source material, i.e. volcanic ashes, have not been investigated in detail and hence, it is highly essential to assess their aptness to be utilized as source material in manufacturing the same because it cannot be generalized to all kinds of precursors. Furthermore, a review of the LCA, i.e., Life Cycle Assessment of Geopolymer composites developed by employing volcanic ashes as the source material is also desirable with an objective to the flashlight on the sustainability of such products and the potential advantages of this novel, innovative Geopolymer technology. Awareness for the promotion of its systematic application should be campaigned among engineers, construction related people, environmentalists, etc. through governments and media especially in the countries suffering from its pessimistic impacts.

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