Replacement of Natural Aggregates in Geopolymer composites - A brief review

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***Abstract:** Natural aggregates are essential constituents of both conventional and geopolymer concrete, which fills up approximately 60–80% of the total volume and have a direct impact on the properties of concrete. The extensive use and the high extraction of the natural aggregates raise questions regarding the conservation of natural aggregates' resources and environments. Therefore, several researchers incorporated some recycled aggregates and waste or by-products (WBP) like copper and steel slag, granite quarry sand, crushed sand, etc. as the substitution of natural aggregates. This literature provides a brief review of the properties of the geopolymer composites when natural aggregates are partially or fully replaced. Several previously published literature has been studied and found that the utilization of recycled aggregates and WBP (to a certain extent) into geopolymer concrete has enhanced physical, mechanical, and durability properties, as well as forming a dense structure. Based on the study, it can be concluded that recycled aggregates and WBP have the potential to partially replace natural aggregates (fine as well as coarse).*

***Keywords:** Geopolymer, Recycled aggregates, Copper slag, Ambient cured, Natural aggregates*.

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1. INTRODUCTION

In the construction industry, Ordinary Portland cement (OPC) is one of the most prominent and commonly used binder substances. India is the world's second-largest producer of OPC, with 298 million tonnes produced annually and accounting for roughly 8% of global OPC production. According to the Bureau of energy efficiency, India, the per capita consumption of OPC is 195 kg, and the demand is continuously increasing. However, the manufacture of OPC has always been controversial because it emits a large amount of CO_2 and demands huge energy [1], [2]. The fact that OPC manufacturing industry emitted approximately 3 billion metric tonnes of CO_2 in 2010, accounting for 5–8% of total CO_2 emissions [3]. Simultaneously, WBP produced by various industries, like fly ash, slag, nano-silica, metakaolin, rice husk ash, silica fume, etc., have increased exponentially and are not managed effectively. To overcome these issues several authors utilized some WBP as the partial replacement of OPC and reported that fresh and hardened properties of conventional concrete (CC) enhanced [4]–[6]. According to Raili Kajaste and Markku Hurme [3] utilization of WBP as the partial substitute of OPC in concrete reduces CO_2 emission by roughly 45%. The inclusion of WBP in concrete does not completely resolve these issues since it can replace the OPC to a certain extent, then it starts reducing the properties of the concrete. Therefore, in the year 1978, professor J. Davidovits introduced Geopolymer as an alternative polymer-based construction material [7].

Geopolymer is an inorganic Al and Si containing polymer with a 3–D amorphous shape and having a chemical structure similar to natural zeolite [8]–[11]. The manufacturing of geopolymer composites decreased energy consumption by 15% and CO_2 emissions by 70% (roughly) compared to the manufacturing of CC [12]. Geopolymer concrete (GC) is most attracted to the construction industry due to its superior or comparable fresh and hardened properties compared to CC by completely replacing OPC with WBP [8], [13]–[15]. The essential constituents of GC are WBP (those are endowed in Al and Si) and sodium or potassium-based hydroxide and silicates solution. According to Turner and Collins [16], the emission of CO_2 from GC was about 9%, which was significantly very small compared to CC. With this Ng et al. [17] documented that manufacturing of geopolymer decreased power requirements by near to 15%. Also, several researchers [8], [13], [17]–[21] reviewed previously published important literature relevant to geopolymer composites and documented that compared to the CC, geopolymer composites had better workability and higher mechanical properties as well as excellent resistance to sulphate, acid, fire, corrosion and freeze-thaw. Geopolymers concrete is one of the best alternatives to conventional concrete. Still, its use is being less in the construction industries of India. Reference [22] highlights the many challenges facing geopolymer composites in India.

The natural aggregates are also the important constituent of CC and GC because they occupy a 60–80% volume of concrete. Ernesto J. Gaudes [9] investigated the effect of coarse aggregate size on the compressive strength of the GC and found that 12.5–25 mm offered maximum compressive strength. Similarly, B. Joseph and G. Mathew [23] explored the behaviour of aggregates on the mechanical properties of GC and informed that GC specimen filled with 70% aggregates (out of which 35% were fine aggregates) provided the highest mechanical properties, like 56 MPa compressive strength, 4.51 MPa split tensile strength and 4.95 flexural strength after 28 days curing. Reference [15] explored the possibility of substituting granulated blast furnace slag (GBFS) sand for river sand in geopolymer concrete. The development of self-cured geopolymer concrete (SCGC) in an environment where temperature and humidity are not controlled has also been attempted. The SCGC had excellent mechanical qualities when it was made up of 60% GBFS sand plus 40% river sand (shown in **Figure 1**). The demand for natural aggregates also increases with the rise of infrastructure, creating an extra burden on the environment by disrupting the ecosystem, because, natural aggregates are mined from riverbeds and rocks, resulting in contamination of the air and water. The widespread consumption and high exploitation of natural aggregate raise concerns about natural aggregate resource management and environmental protection. To preserve the natural aggregates, several researchers [7], [24]–[30] used recycled aggregates and WBP to replace natural aggregates and concluded that the mechanical properties of GC or geopolymer mortar (GM) were enhanced with the inclusion of these aggregates, however, the angular or irregular particle size of recycled aggregates and WBP deteriorated the flow and workability. This literature offered a brief review of the investigations conducted by several researchers using recycled aggregates and WBP as the substitution of natural aggregates (fine aggregates as well as coarse aggregates).

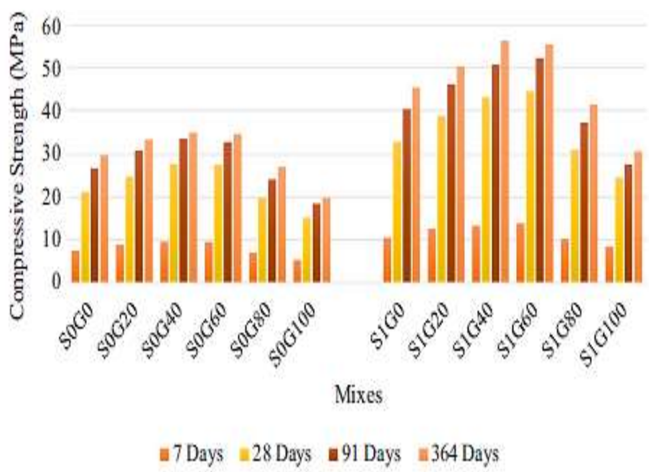

Figure 1. Effect of GBFS sand on the compressive strength of various SCGC mixes [15].

2. UTILIZATION OF THE RECYCLED AGGREGATES AND WBP

The incorporation of recycled aggregates and different WBP such as copper slag, granulated smelter slag, steel slag, ferronickel slag, manufactured sand, granite quarry sand, etc. have a different effect on fresh and hardened properties of prepared GC/GM due to their varying characteristics. Moreover, high-temperature curing and ambient curing also highly affect the properties of concrete. Consequently, this section is further classified into two sections: the first section will discuss the effect on heat-cured GC/GM and the second will discuss the effect on ambient GC/GM.

2.1 Heat-cured geopolymer concrete/mortar

Recycled aggregates are widely used material as a replacement for natural aggregates because of their easy availability in large quantities and the lack of additional work needed. J. Xie et al. [26] used recycled aggregates (taken from the concrete of demolished structures with the compressive strength of 20–30 MPa) as a complete replacement of natural coarse aggregates. The prepared concrete specimens have been cured at 80 ^{0}C temperature for 24 h and tested on the seventh day. It was concluded that incorporation of ground granulated blast furnace slag (GGBFS) into the fly ash-based GC (with recycled aggregates), contributed superior physical and mechanical properties to GC as compared to that of CC. The manufacturing of the C–S–H gel along with the C–A–S–H and N–A–S–H gel, and a strong bond formation between the GGBFS and fly ash-based geopolymer matrix and recycled aggregates could be attributed to the enhancement of the properties of GC. Results also informed that to achieve superior properties of GC with recycled aggregates, GGBFS must be added as the substitution of fly ash. In a different study, J. Xie et al. [31] added that the inclusion of metakaolin as the substitution of GGBFS further enhanced the properties of GC. Similarly, P. Nuaklong et al. [32] associated 4.5–9.5 mm size crushed limestone and recycled aggregates as the alternative to natural coarse aggregates into the high calcium fly ash and metakaolin based GC, and reported that GC with recycled aggregates exhibited lower mechanical and durability properties than GC with limestone, however, slump increased. The presence of absorbed water within the recycled aggregates may have influenced the properties of GC by taking part in the geopolymerization reaction (GPR). Metakaolin creates a dense microstructure and speeds up the geopolymerization process, resulting in better mechanical properties and a shorter setting time [31], [32] compared to that of fly ash based GC. In another study, P. Nuaklong et al. [33] investigated the influence of recycled aggregates as fine aggregates in the high-calcium and low-calcium fly ash based geopolymer mortar and concluded that degradation in the compressive strength was found with the increase of recycled aggregates. They recommended that geopolymer mortar containing recycled aggregates not be utilised in a harsh environment (shown in **Figure 2**).

P. Posi et al. [34] attempted to produce lightweight GC by substituting recycled aggregates obtained from lightweight blocks for all types of natural aggregates (fine aggregates as well as coarse aggregates). The molarity of sodium hydroxide was 5, 10 and 15 M, and particle size of recycled aggregates was varying in between 0.001–1.18, 1.18–4.75 and 4.75–12.5 mm. Experimental results informed that the production of lightweight GC with a density of 860–1400 kg/m^3 and compressive strength of 1.0–16.0 MPa is possible by using recycled aggregates. Researchers [35] used recycled ceramic waste as an alternative to fine aggregates in heat-cured GC. They found that using RCW as a fine aggregate reduces the resistivity against acids and the strength of GC. V. Sata et al. [36] utilized two different types of recycled aggregates, one was collected from structural concrete members and the other was collected from clay bricks, as the natural coarse aggregates for developing the pervious GC, and concluded that both types of aggregates provided less mechanical properties compared to GC with natural coarse aggregates. Likewise, reduction in mechanical and durability properties was observed by F.U.A. Shaikh [37], when recycled aggregates has been utilised as coarse aggregates and the sample was cured in steam at 60 ^{0}C for 24 h. It may be due to the presence of the micro-cracks in recycled aggregates, which form a weak bond between the aggregate and geopolymer paste. In this manner, A. Wongsa et al. [38] utilized the bottom ash as the substitution of natural fine aggregates and coarse aggregates and attempted to develop a lightweight GC. It has been observed that mechanical and durability properties were reduced compared to that of natural aggregates, and these values further decreased with the increment of sodium silicate/sodium hydroxide and liquid/ash ratio.

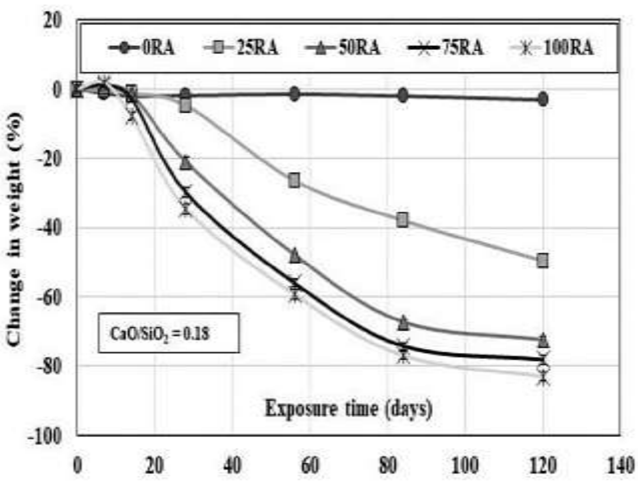

Figure 2. Change in weight (%) in geopolymer mortar specimens immersed in acid solution [33].

Manufactured sand and quarry dust are generated as waste material after crushing granite stone. Such aggregates are commonly used as an alternative to natural fine aggregates. Saravanan. S et al. [7] investigated the effect of manufactured sand on the compressive strength of low calcium fly ash based GC, which was cured at 60 ^{0}C for 24 h, and reported that around 10% compressive strength increased when natural fine aggregates were completely substituted by manufactured sand. Furthermore, raising the molarity of sodium hydroxide by 10 to 12 M boosts compressive strength by 8%. Iftekhair Ibnul Bashar et al. [39] utilized manufactured sand and quarry dust as a replacement of conventional mining sand in the palm oil fuel ash (POFA) based GM and investigated the effect on the workability, density and compressive strength. As a consequence of the findings, it has been ascertained that the highest compressive strength has been achieved by 100% quarry dust compared to that of other replacement percentages of manufactured sand and quarry dust. Moreover, 100% manufactured sand achieved approximate similar strength to the palm oil fuel ash based GM with conventional mining sand. In another study, Iftekhair Ibnul Bashar et al. [40] informed that a mixture of 50% POFA and 50% GGBFS can be used as a binder material in GC with manufactured sand (as an alternative to fine aggregates), and could achieve 56 MPa compressive strength.

K. Mermerdaş et al. [25] considered four different grades such as 0–4, 2–4, 1–2, and 0–1 mm of crushed limestone as a replacement of natural river sand in GM (those were initially cured for 24 h at 90 ^{0}C temperature), and investigate the effect on fresh properties like workability and fresh density, and hardened properties such as compressive strength, split tensile strength, water absorption and water sorptivity. It was observed that due to the inclusion of crushed limestone fresh properties of GM decreased, however, hardened properties enhanced. It has been also observed that the 2–4 mm grade achieved the highest mechanical properties just after one day of casting, however, the 0–4 mm grade showed less water absorption and water sorptivity compared to the other grade. M. Albitar et al. [27] investigated the influence of granulated lead smelter slag (GLSS) as an alternative to the binder and fine aggregates on the compressive strength of fly ash based GC. **Figure 3** showed that GLSS as a complete replacement of natural river sand did not show a significant effect on the compressive strength of the only fly ash based GC, however, a significant improvement on compressive strength was observed when GLSS substituted 50% fly ash, which signifies that GLSS also improved the GPR. Furthermore, the reduction of fly ash into the GC with 100% GLSS (as fine aggregates), reduced the compressive strength. It may be due to the unavailability of aluminosilicate compounds for completing the GPR and poor reactivity of GLSS.

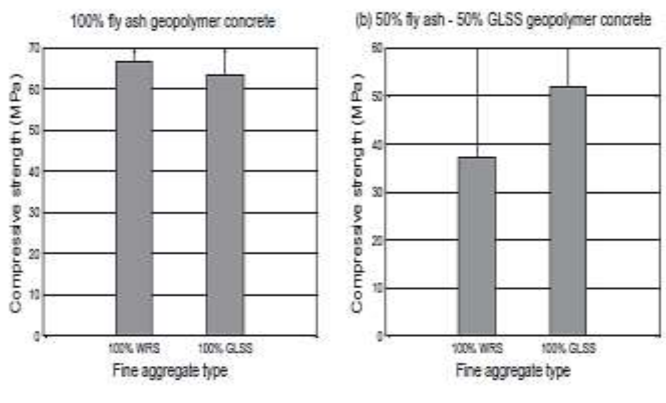

Figure 3. Influence of GLSS as fine aggregate on compressive strength [27].

M.S.H. Khan et al. [29] examined the behaviour of steel furnace slag (SFS) as an alternative to the natural coarse aggregates in 90% low calcium fly ash and 10% GGBFS based GC on the physical and mechanical properties. As compared to GC with natural aggregates, the compressive strength, surface resistivity, and pulse velocity of the GC with SFS were higher, however, no significant effect has been observed on the shrinkage and expansion of the GC specimens. It was found from the microstructural study that the inclusion of CaO and MgO in GC from the SFS aggregates, reduces the cause of delayed expansion. A.M. Rashad et al. [41] investigated the possibility of granulated blast furnace slag (GBFS) as the full or partial replacement of natural fine aggregates in alkali-activated slag (AAS) mortar and evaluate the effect of compressive strength after the sample was exposed to 200, 400, 600 and 800 ^{0}C for 2 h. An increment in the residual compressive strength and reduction in the formation of microcracks were observed, with the increment in the quantity of GBFS, which signifies that GBFS as fine aggregates can resist high temperature and could be used as an alternative to fine aggregates (**Figure 4**). A dense interfacial transition zone (ITZ) has been observed, which signifies the formation of a strong bond between SFS or GBFS aggregates and geopolymer binder probably the cause for the enhancement of compressive strength [29], [41].

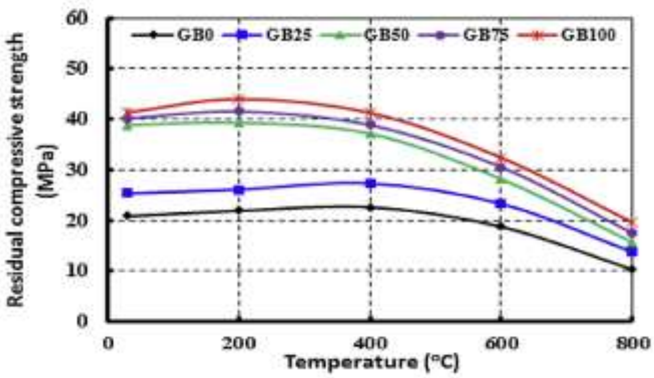

Figure 4. Residual compressive strength (MPa) of AAS mortar [41].

2.2 Ambient cured geopolymer concrete/mortar

Based on previous research, it appears that less research has been done to evaluate the impact of alternative aggregates on the ambient cured geopolymer concrete. C. Sreenivasulu [42] incorporated the granite slurry as the replacement of natural fine aggregates in ambient cured fly ash and GGBFS based GC. Short-term mechanical properties were enhanced by using granite slurry to replace fine aggregates up to 40%, further increment in the quantity of granite slurry showed a negative effect. The explanation for this behaviour may be that granite slurry (up to 40% replacement of fine aggregates) acts as a filler material, forming a dense ITZ and enhancing the mechanical properties, however, after 40%, the quantity of finer particles increased in the geopolymer matrix, which may hinder the GPR. Similarly, X. Cong and W. Zhou [30] investigated the influence of water-quenched slag as substitution of fine aggregates in the GGBFS and fly ash based AAS mortar and found that the flowability, shrinkage and setting time declined. Micro grains of 40–200 µm in diameter were formed during the water quenching phase of slag as illustrated in **Figure 5**, as well as water quenched slag having irregular particle size, rough surface area, and pore structure, requiring more liquid to form a uniform paste. As a result, flowability, shrinkage, and setting time were all greatly influenced. However, these micro grains were involved in the GPR later stage, which improved the mechanical properties.

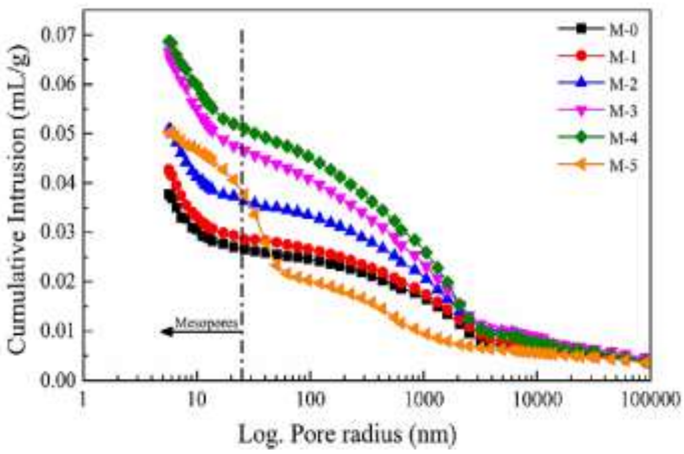

Figure 5. Change in pore size after 90 days of curing [30].

K. Parthiban and K. Saravana Raja Mohan [43] examine the effect of recycled aggregates (as partial or full replacement of coarse aggregates) on the workability, mechanical and durability properties of the AAS concrete. It was found that the workability of AAS concrete continuously decreased as the recycled aggregates content increased, because of the creation of voids with the inclusion of recycled aggregates, whereas, it can be avoided by the inclusion of a superplasticizer. The mechanical and durability properties of AAS concrete with only recycled aggregates were higher than those of CC with natural coarse aggregates, but AAS concrete with 50% recycled aggregates performed better than the other AAS concrete. The fact that the quantity of fine particles increases with the inclusion of recycled aggregates, reduced the alkali solution to complete the GPR and decrease the mechanical properties. Similar results were also observed by X. Ren and L. Zhang [44] and informed that recycled aggregates have significant potential to replace coarse aggregates.

A.M. Aly et al. [45] used crumb rubber as the substitution of both types of natural aggregates (fine aggregates as well as coarse aggregates) and studied the hardened properties and impact resistance of GGBFS based and water cured GC. Results showed that crumb rubber had a negative effect on the hardened properties, whereas, impact resistance and ductility of GC enhanced. Moreover, it has been suggested by the researchers that GC with crumb rubber can be replaced CC, particularly at the roads, runway and taxiway. Similarly, F. Aslani et al. [46] also reported that the addition of crumb rubber in lightweight GC decreased the mechanical properties, which can be explained by the fact that crumb rubber as aggregates have more air voids, lower density and create weak ITZ bond compared to the natural aggregates. Z Yahya et al. [47] understand the durability properties of GC with crumb rubber after immersing the specimens in seawater and found that the weight of the samples increased as the exposure time and the amount of rubber increased, which can be seen in **Figure 6**.

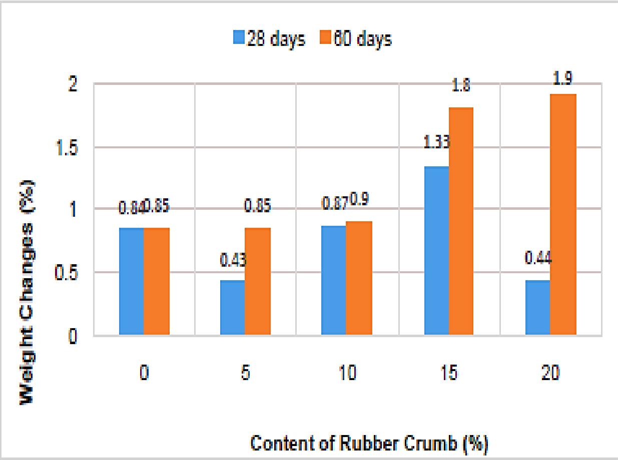

Figure 6. Change in weight after 28 days and 60 days of exposure [47].

To investigate the durability properties of GC, B. Parthiban and S. Thirugnanasambandam [48] incorporated recycled waste glass (RWG) as the full replacement for fine aggregates and studied the compressive strength, chloride permeability, and pull-out strength, along with the effect of acid and sulphate on the weight and compressive strength. In contrast to GC with natural fine aggregates, GC with RWG exhibited greater resistance to acid, sulphate, and chloride, this may be attributed to lower water absorption of RWG. B. Mithun and M.C. Narasimhan [49] and C. Sreenivasulu et al. [50] explored the effect of copper slag as an alternative to natural fine aggregates on the performance of AAS concrete and fly ash and GGBFS based GC, respectively. It was observed, copper slag has amorphous nature that develops a uniform and dense ITZ and magnified the alkali-activation and GPR, by which the performance of AAS concrete and GC were enhanced. Moreover, **Figure 7** informed that the performance of AAS concrete was much superior to the CC when specimens were exposed to the acidic environment [49].

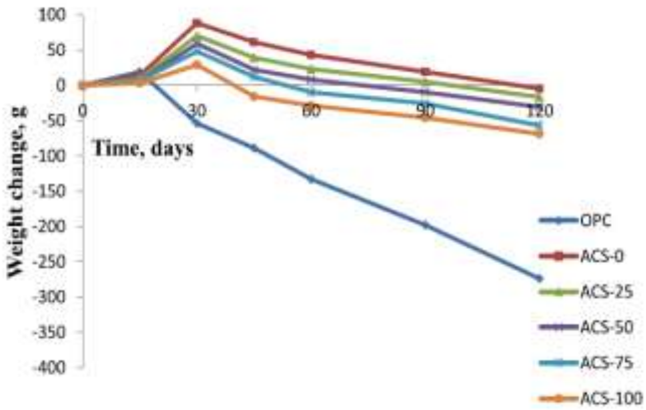

Figure 7. Change in weight in the various specimens after exposure to the acid solution.

3. CONCLUSIONS

The manufacturing of OPC and natural aggregates requires high energy, pollute the environment and imbalance the ecosystem. Along with this problem, the production of recycled aggregates and WBP in high quantity is also creating the problem. Therefore, the determination of some alternative for OPC and natural aggregate is the need for this hour. The GC is the best alternative to CC, to date, which utilizes fly ash as the full replacement to OPC. However, to achieve high strength GC required high-temperature curing, which is an obstacle to using GC for cast-in-situ. Therefore, some studies incorporated WBP (those are rich in calcium) as the partial or full replacement of fly ash. Whereas, researchers still finding the best alternative to natural aggregates, therefore, if recycled aggregates and WBP are used as replacements for natural aggregates, then it is ice on the cake. This study evaluated the efficacy of the recycled aggregates and different WBP as the substitution of natural aggregates, in the high-temperature, steam, ambient and water cured GC. From the above discussion following conclusions can be drawn –

- Recycled aggregates have the potential to partially replace natural coarse aggregates of the heat-cured or ambient cured geopolymer concrete.
- Many WBP like manufactured sand, copper and steel slag, etc. have the potential to partially replace natural fine aggregates.
- The properties of recycled aggregates and WBP aggregates, such as particle shape and size, roughness, water absorption, density, etc. highly influenced the properties of GC.
- The utilization of a small quantity of crumb rubber in GC enhances the ductility and impact resistance of GC.
- Since natural aggregates do not highly participate in the GPR, whereas, the chemical composition of WBP affect the properties of GC by magnifying the GPR.
- Most of the substitutes, form dense microstructure and create a stronger bond with the binder which may be the cause for the enhancement of properties of GC.

- The fully or partial replacement of aluminosilicate source material in GC has further enhanced the properties of GC with recycled aggregates or WBP.

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