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GEO-POLYMERIZATION OF RECYCLED CONCRETE AGGREGATE- A REVIEW

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Abstract- *Worldwide concrete production is increased due to massive infrastructure developments and increasing world population. To meet the rising demand of concrete, leads to production of huge quantity of ordinary Portland cement (OPC) and utilization of enormous amount of natural aggregates. The production of OPC liberates huge amount of CO_2 to the atmosphere which leads to the global warming. The rising demand of environment friendly construction materials has been the driving force for developing sustainable and cost-effective construction materials. Geo polymer an alternative binder to OPC is gaining increased interest with lesser CO_2 emission in comparison to OPC. An ample summary report on the outcomes of extensive literatures reveals the benefits of incorporating recycled concrete aggregate (RCA) in geo polymer concrete (GPC) mixtures, constituents of GPC, strength and its potential applications in various construction fields. In near future, GPC needs further research to make it as a user friendly construction material for practical applications in construction sector.*

Keywords-*Sustainable concrete; Recycled aggregate; Geo polymer concrete; Fly ash; Sodium silicate; Sodium hydroxide*

I. INTRODUCTION

Globally, Concrete is the second most consumed premier construction material after water and it shapes the built environment. It is the versatile material made up of cement, aggregates, water and admixtures or partial cement replacement materials that can be easily mixed to meet a variety of special needs, formed to virtually any shape and size. The quality of concrete is depends upon the quality and quantity of the ingredients. The consumption of OPC causes pollution to the environment due to the emission of CO_2 [1], [2],[3].The contribution of cement production worldwide to the greenhouse gas emission is estimated to be about 1.35 billion tons annually or about 7% of the total greenhouse gas emissions to the earth's atmosphere [4].The global demand of the OPC will have increased by almost 200% by 2050 from 2010 levels [5], and hence the impact of the OPC industry on the environment is believed to become a real issue. With the aim of reducing the production of OPC, a new type of green and environment-friendly concrete, namely, GPC has recently gained some research attention.

Therefore, the introduction of a novel binder called 'geo polymer' by Davidovits promises a good prospect for application in the concrete industry as an alternative binder replaces cement completely and reduces the environmental deterioration. The concrete consists of 70-75% of the aggregates by volume and plays a vital role in various fresh and hardened properties of concrete such as workability, mechanical strength and durability. It is obtained from natural resources; the demands for natural resources are more than the availability. The construction industry is one of the economic sector that are more responsible for consuming enormous amount of natural resources and generating waste materials. The management of construction and demolition waste (C and D) also creates a problem. The lack of land available for the disposal and expenditure of C and D waste is becoming difficult and higher. This has led to the promotion of waste recycling as a measure to reduce waste and to mitigate the detrimental effects of construction activities on the environment [6].

The use of recycled concrete aggregate (RCA) has gained remarkable drive in constructional engineering. Utilization of recycled waste concrete would benefit into two folds. First, lower the environmental pollution, reduction in valuable landfill space, and savings in natural aggregate resources [7].Due to environmental and technical factors, researchers around the world are trying to discover a ways to reuse these materials effectively. [8], [9],[10],[11], [12], [13], [14].On the other hand, coal burning power generation plants produce huge quantities of fly ash. The volume of fly ash would increase as the demand for power increases. Most of the fly ash is considered as a waste and dumped in landfills. Fly ash based geo polymer also provided better resistance against aggressive environment and elevated temperature compared to normal concrete. In addition to providing a gainful use for waste materials and reducing green house gases, the geo polymerisation has also been shown to immobilize some heavy metals present in the fly ash [15].The production of high strength concrete by using fly ash as a source material for geo polymerization of RCA for structural applications gains importance for environmental sustainability. This eliminates the need for the production of OPC and use of construction waste as a constituent in new concrete. Development of geo polymer as alternative cement-less binder to OPC was one of the significant breakthroughs in the field of concrete technology in the 20th century [16] and also it can provide a solution to produce greener concrete for sustainable development.

II. GEOPOLYMER CONCRETE

The term 'geo-polymer' was first introduced by Davidovits in1970s to represent the mineral polymers resulting from geochemistry, as alternative building materials. Geo-polymer is a promising alternative binder to OPC for making concrete. There are two chief constituents of GPC namely the source material and the alkaline solution.

The Figure: 1 represents the constituents of GPC. The source material rich in silicate and alumina reacts with alkaline solution to produce alumino silicate gel that binds the aggregate to produce a good concrete [17]. The production of one tons of geo polymer binder generates 0.18 tons of CO_2, from the combustion of carbon-fuel, compared to one tons of CO_2 from OPC production [18].

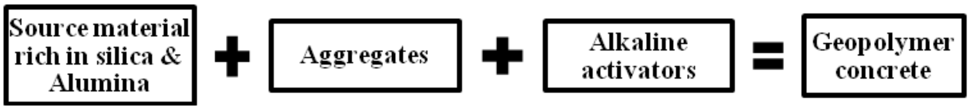

Figure: 1 constituents of geopolymer concrete

2.1. Source Material

The strength of geo polymer depends on the nature of source materials. GPC is produced by utilizing industrial by-product materials such as fly ash, ground granulated blast furnace slag (GGBS), and other alumino-silicate materials originated from geological sources such as meta kaolin as a source material; hence produces low CO_2 in comparison to OPC [19].Fly ash is a by-product from the thermal power stations, which is abundantly available worldwide, but to date its utilization in construction sector is limited. Moreover, the use of fly ash is more environmental friendly and save cost compared to OPC. In India, at present nearly 90 metric tons of fly ash is generated per annum and is largely responsible for the environmental pollution, only 3% is being consumed [20]. GPC utilizing fly ash as a source material has been most broadly studied due to the environmental advantages, its abundance, and high silicon and alumina content. ASTM classifies the fly ash into two types according to the type of coal used for burning in thermal power stations. The chief difference between these types is the amount of calcium, Silica, alumina and iron content in the fly-ash. The class F fly ash produced by burning Anthracite and bituminous coal. Class C fly ash is produced by burning lignite or sub-bituminous coal. Class C fly ash has self-cementing properties which is relatively rich in CaO, tends to set far too quick to be used in any practical application [21].Additionally the high amounts of CaO may lead to durability issues in GPC made from Class C fly ash [22], [23]. GPC made from fly ash and ground GGBS results in lower CO_2 emission than OPC concrete. Meta kaolin based GPC having low Si /Al ratio due to this high amount of sodium silicate is required causing a high environmental impact [24]. From the Figure: 2, Predominantly all over the world approximately 90% of the researchers utilize fly ash and only 10% of the researchers utilize slag and Meta kaolin as a source material to produce high strength GPC economically. Geo polymers made from calcined source materials, such as Meta kaolin (calcined kaolin), fly ash, slag etc., yield higher compressive strength when compared to those synthesised from non-calcined materials, such as kaolin clay. The source material used for geo polymerization can be a single material or a combination of several types of materials [25]. The choice of the source materials for making geopolymers depends on factors such as availability, cost, and type of application [26].

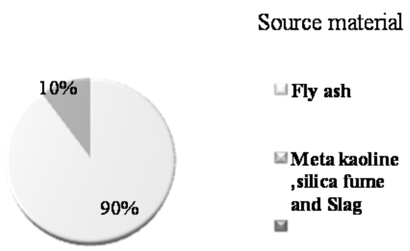

Figure.2 Utilization rate of source material

2.2. Alkaline activators

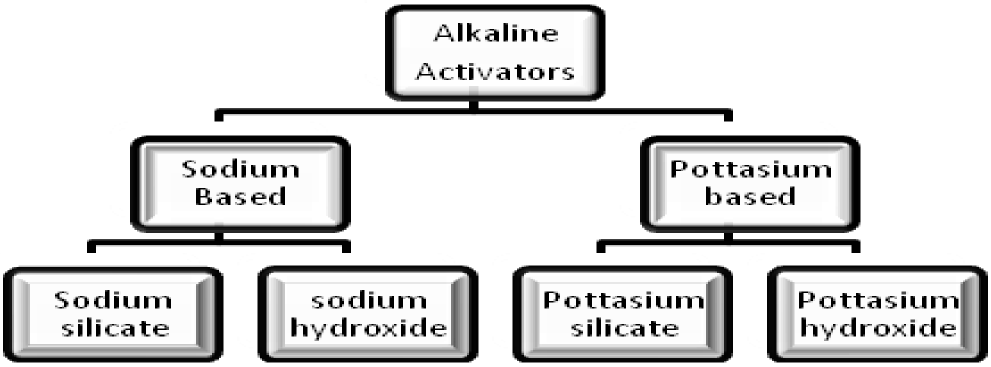

Figure: 3 Alkaline activators

The nature of alkaline liquid plays an important role in the polymerization process. Reactions occur at a high rate when the alkaline liquid contains soluble silicate, either sodium or potassium silicate, compared to the use of only alkaline hydroxides [27]. The most common alkaline activators used in geo polymerization shows in a Figure: 3. it is a combination of sodium hydroxide (NaOH) or potassium hydroxide (KOH) and sodium silicate or potassium silicate [28]. Sodium based solutions were chosen because they were cheaper and it liberates silicate and aluminate monomers .The high concentration of NaOH solution will improved the compressive strength properties of GPC produced because the higher concentration of NaOH creates the good bonding between aggregate and paste of the concrete [29].

III. RECYCLED CONCRETE AGGREGATE (RCA)

The use of recycled aggregates (RA) obtained from C and D waste is feasible for the manufacture of recycled structural concretes. Even though, applications in which the recycled concrete is subjected to important cyclic loading, the substitution of 100% of the recycled coarse aggregate is not recommended [30].
Although the role of aggregates in GPC is similar to that in OPC, the binder in GPC is formed by the reaction of silica and alumina rich material with alkaline liquids and has not been linked to any hydration process [31].As the aggregates have the same role in both OPC and GPC mixes, it is therefore reasonable to consider and employ the same methods for selecting aggregate proportions in GPC mixes as usually adopted for OPC [32]. GPC containing smaller size coarse aggregates exhibited slightly higher compressive strength at all elevated temperatures than those containing larger coarse aggregates, because of the possibility of less micro cracking in the interfacial transition zone (ITZ) of aggregates in the former [33]. Geo polymer seems to have the potential to produce high Performance Concrete based on high volume RA. They can achieve a very high compressive strength just after one or two days, allowing for shorter concrete structures execution [34].

IV. MIX DESIGN AND MIXING METHOD

GPC is a new type of construction material due to this standard mix design approaches are not yet available. The constituent of GPC is heterogeneous in nature, whose interactions and final structure and chemical composition are under intensive research. Therefore, the formulation of the GPC has to be done by trial and error basis [35]. In the design of GPC mix, coarse and fine aggregates together were taken as 77% of entire mixture by mass. This value is similar to that used in OPC concrete in which it will be in the range of 75 to 80% of the entire mixture by mass. Fine aggregate was taken as 30% of the total aggregates. The density of GPC is taken similar to that of OPC as 2400 kg/m^3 [27].Alkaline activator liquids are prepared at required ratio 24 hrs prior to casting to neutralize the heat liberated from the alkaline liquid. The source material rich in alumina and silica is mixed with both fine and coarse aggregates in dry condition. Finally alkaline solution is mixed in to the dry mixture. The super plasticizer is used to maintain the workability of GPC.

V. FACTORS INFLUENCING THE GEOPOLYMERISATION

5.1 Silicates to hydroxide ratio

Higher silicates to hydroxide ratio result in higher strength, but also increase the unit cost of the GPC system [36], [32]. Higher Na_2SiO_3 to NaOH ratio generally reduced the workability of the fresh mixtures due to the higher viscosity of Na_2SiO_3. While higher concentration of NaOH was found to increases the setting time of the resultant geo polymer mixture [37]. The ratio was varied from 2 to 3.5, in the increment of 0.5. The average maximum strength was obtained when the Na_2SiO_3 to NaOH ratio was 2.5 [38], Joshi et al 2012, Bakri et al 2013, [44].

5.2 Molarity of NaoH

Molarity of NaOH solution plays a vital role in the formation of strength in GPC. The mass of NaOH solids in a solution depends on the Molarity. Generally, the molarity of NaOH may taken from 6M to 18 M. The GPCs containing high molar NaOH solutions produces higher compressive strength. This is due to the higher concentration of NaOH will make the good bonding between aggregate and the paste of the concrete (Ismail et al 2011, Carlos et al 2012). GPC with 12M of NaOH concentration produced optimum compressive strength at 2, 3 and 7 days of testing (Bakri et al 2011, [36]. The higher concentration of NaOH may increase the cost of the construction.

5.3 Water to geopolymer solid ratio (W: GPS)

This is the ratio of the total mass of water in the system, including water in the hydroxide, silicate solutions and any additional free water, to the sum of the mass of fly ash, alkaline hydroxide and silicates solids. As these solids contribute to the 'geopolymerisation' process they are termed as geopolymer solids. It has been proposed that this ratio is analogous to the water to binder ratio in OPC with similar effects on the GPC mix [27].The compressive strength of GPC increases as the total water to geopolymer solid ratio decreases [27], Lloyd 2010, Olivia et al 2011, [43].

5.4 Alkaline liquid to fly ash ratio (AL: FA)

This is the second important ratio in the mix design of GPC. The compressive strength increases when AL: FA increases. The alkaline liquid component is the sum of the masses of Na_2SiO_3 and NaOH solution. The fly ash element is simply the mass of fly ash. The most previous research has used AL: FA ratios in the range 0.3-0.5 [35].
When the AL: FA ratio is increased above a certain level, then the compressive strength will actually decrease, instead of continuing to increase [38]. The reason for this strength loss at higher ratios of AL: FA is, the fly ash particles, with a finite surface area, are saturated with alkaline liquids at a certain point, and any more alkaline liquid is simply wasted within the mix. This excess alkaline liquid effectively then becomes extra water, diluting the mix and affecting the bonding between aggregate and binder. The lowest potential ratio is to be 0.50. Higher alkaline liquid to fly ash ratio generally resulted in higher workability of the fresh geo polymer mixture.

5.5 Water for Mixing

Water in GPC mix does not take any part in the chemical reaction. In OPC concrete, water in the mix chemically reacts with the cement to produce a paste that binds the aggregates. The chemical reaction occurs in geo polymers produces water that is eventually expelled from the binder. It has been observed that additional water content in the GPC mixture affected the properties of concrete in the fresh state as well as in the hardened state. The compressive strength of GPC decreases as the water content increases [35].This is due to the formation of weak polymerisation products in presence of extra water. Similar to OPC concrete, the use of excessive water in geo polymer concrete also adversely affects its strength and durability properties, because of the formation of excessive voids through evaporation of some unbound water during heat curing [33].

5.6 Addition of Super plasticiser

In fresh state, the GPC has a stiff consistency. Although adequate compaction was achievable, an improvement in the workability was considered as desirable. As the concentration of super plasticizer increases the amount of water required decreases. In order to improve the workability of fresh concrete, high-range water-reducing naphthalene based super plasticiser was added to the mixture. The dosage of super plasticizer also has an effect on the compressive strength of the concrete. Concrete mix with 2% dosage of super plasticiser has achieved higher compressive strength as compared to the 3% and 4% dosage of the super plasticiser. There is a slight degradation in the compressive strength of hardened concrete when the super plasticizer dosage is greater than 2% by mass of source material [39], [36]

5.7 Rest period prior to curing

The term 'Rest Period' is defined by the time taken from the completion of casting of concrete specimen to the start of curing at an elevated temperature. One day rest period increases the compressive strength of fly ash based GPC as compared to that without rest period. The start of the heat curing may be delayed for several days without adversely affecting the properties of GPC. In fact, an increase in the rest period significantly increases the strength of the final product [27], [39].

5.8 Effect of curing methods on strength

Curing method is an important factor for the strength point of view of GPC. The figure: 4 show the different curing methods available for the production of high strength GPC.

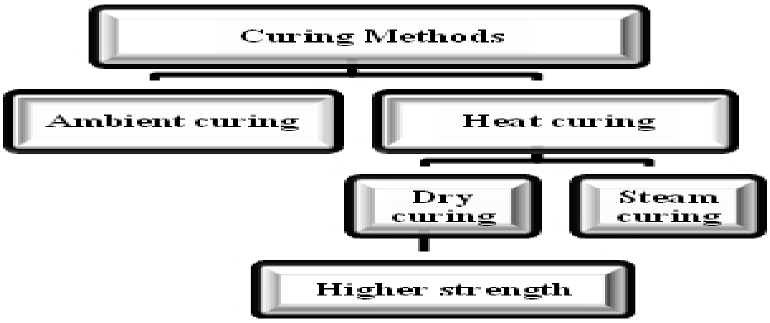

Figure: 4 Curing Methods

However, curing at room temperature has successfully been carried out by using calcined source material of pure geological origin, such as Meta kaolin [25], [40].Although GPC can set at ambient temperatures; application of heat to the specimen greatly increases the compressive strength. Heat curing can be achieved by either steam curing or dry curing. The compressive strength of dry cured GPC is approximately 15% larger than that of steam cured GPC [35]. Dry curing results in higher strength when compared to wet curing at similar temperatures; strength increases as the time and temperature of curing is increases [32].The behaviour of GPC at elevated temperatures affects by the size of the aggregates. The aggregate with smaller sized (<10 mm) could lead to spalling and also extensive cracking of geo polymer but the larger aggregate (>10 mm) were more stable [41].

5.9 Curing time & temperature

The main polymerization process or the chemical reaction of GPC takes place with the temperature imposed to it during curing. The temperatures of curing as well as the curing time play a significant role in the final strength of GPC mix [42].The process of curing and hardening is different for GPC than the OPC.GPC tend to increase the strength when curing at elevated temperatures, ranging from room temperature to nearly 100°C depends on the source material used [15]. The compressive strength of GPC increases with increase in curing temperature up to a value of 100 °C and beyond which it decreases [43], [44]. Due to the increase in temperature, polymerization becomes more rapid, and the concrete can gain 70% of its strength within 3 to 4 hours of curing [41].The loss of strength beyond the curing temperature of 100°C is due to the loss of moisture from the specimen. Even if sealed properly, the specimen may dry out and lead to reduction in strength [43].

Table: 1 Effect of curing regime on compressive strength

Author	**Curing regime**	**Compressive strength**	**Findings**
Ozbakkaloglu et al 2015	Ambient temperature.	34.3Mpa @ 28 days	Ambient temperature tends to develop lower Compressive strength than OPC.
Sarker et al 2014	60°C oven curing 24 hrs. Ambient curing 22° C	55MPa @ 28 days 40 MPa @ 28 days	Ambient curing leads to lower the compressive strength.
Rangan, 2005	60°C oven curing, curing time varied from 4 hrs to 96 hrs (4 days).	82 Mpa @ 7 days.	Longer curing time improved the polymerization process resulting in higher compressive strength.
Nasir Shafiq, 2011	70°C oven curing for 24 hrs	53.80 Mpa @ 28 days	An increase in curing temperature beyond 70°C did not increase the strength Substantially.
Hardjito et al 2008	90°C oven curing for 48 hrs	70Mpa @ 7 days	Higher curing temperature leads to higher compressive strength.
Prakash et al 2013	75°C oven curing for 24 hrs. 90°C oven curing for 24 hrs.	46 Mpa @ 7 days 49 Mpa @ 7 days	Increase in curing temperature beyond 75°C has yielded into minor increment in the compressive strength for the concrete.

The effect of curing regime on compressive strength is illustrated in Table 1: A lower degree of geo polymerization experienced by GPC at ambient temperature curing which results in an insignificant exothermic reaction of the GPC [45], [35]. Presence of sodium silicate as a gel form takes delay in setting at ambient temperature. In order to overcome this, curing at elevated temperature is recommended which leads to increase in compressive strength [46]. Longer curing time improved the polymerisation process resulting in higher compressive strength and did not decrease the compressive strength of GPC [36], [40].

VI.PROPERTIES OF GEOPOLYMER CONCRETE

Geo polymer also exhibit similar or superior engineering properties compared to OPC. The compressive strength of geo polymer predominantly depends on the content of fineness of the fly ash particles, which smaller than 43µm would be more active in alkali-activation process. This is due to the finer particles have higher amorphous content which can get more chances to react with solution which lead to higher strength [47]. Low calcium fly ash based GPC has been reported to have excellent compressive strength, resistance to sulphate attack, and good acid resistance [22]. Setting time and compressive strength of geo polymers varied with the variation of alkaline activator to fly ash ratio and sodium silicate to sodium hydroxide ratio in the alkaline activator solution [48]. An increase in strength and decrease in setting time was observed with the increase of CaO content in the source material [21]. The 90% of the final compressive strength is reached within 7 days in case of GPC, and there is not much variation in compressive strength after 7 days [49]. For all ratios of fly ash to alkaline activator, compressive strength decreased at Na_2SiO_3 to NaOH ratio of 3.0. This might be due to excess of OH^- concentration. In addition, the excess sodium content may also form sodium carbonate due to atmospheric carbonation and may disrupt the polymerization process [50].The unsealed specimens exhibited higher 7-day compressive strength than those of the sealed specimens. This observation can be attributed to that the sealed curing prevented the evaporation of water from the GPC, and the presence of excess water in turn resulted in a lower compressive strength [45].

Works on reinforced GPC are not many and however, the existing test results shows that structural behavior of GPC and conventional concrete are similar in nature, except that sometime at the same strength level, GPC may tend to have lower modulus of elasticity [51]. The shrinkage strains and associated weight losses of the GPC were higher than that of the OPC. This is due to the lower degree of geo polymerization experienced by GPC at ambient temperature, which resulted in structures containing unreacted porous coal ashes. Heat cured fly ash based GPC undergoes very low drying shrinkage [22].The unreacted coal ashes were filled with water, which was subsequently released during the chemical reaction process of geo polymer. In ambient curing conditions, this water evaporated over a period of time resulting in higher drying shrinkage of GPC [45]. Geo polymer composites have performed better than OPC composites in durability related tests such as sulphate, acid, fire and corrosion resistance. This is mainly due to polymeric nature of GPC matrix without presence of free lime [51].

VII.POTENTIAL OF GEOPOLYMERIC BINDER IN RECYCLED AGGREGATE CONCRETE (RAC)

Several studies have been carried out to reduce the use of OPC in concrete to reduce the carbon foot print and to address the global warming issues. RAC needs more amount of cement to eliminate the inferior properties of RA and to attain high strength. Cement is conventionally used as the primary binder to produce concrete.

Geopolymers are formed by alkaline activation of an alumino-silicate material like fly ash, Meta kaoline, Rice Husk Ash, activated bentonite, clay and red mud etc. Effective use of fly ash in GPC further reduces the environmental pollution caused by the fly ash [43]. In geopolymeric recycled aggregate concrete (GRC), the reaction products are mainly amorphous aluminosilicate gels. Moreover many fly ash particles were embedded well in paste and fill up the pores. Comparatively, the paste of GRC is denser and homogeneous, which leads to higher strength [11]. Geo-polymers seem to have the potential to produce high performance concrete based on high volume recycled aggregates (RAs). They can achieve a very high compressive strength just after one or two days. They also have high resistance to acid, sulphate, chloride, heat and fire. Geopolymers have the remarkable capacity to immobilize harmful constituents which is a crucial feature when contaminated RAs were used [11],[34].

VIII. CONCLUDING REMARKS

To date; several studies have been carried out to minimize the use of OPC in concrete to address the global warming issues. The following conclusions were drawn from the extensive literature study to reduce carbon footprint of the construction industry.

1. Higher the fineness of fly ash particles gives a higher compressive strength because of more surface area with more Si-Al bond for polymerization. Fly ash based geopolymer is better than normal concrete in many aspects such as compressive strength, exposure to aggressive environment, workability and exposure to high temperature.

2. The optimum strength was obtained when the Na_2SiO_3 to NaOH ratio was 2.5. The incorporation of RAs in GPC becomes a great alternative to natural aggregates. The use of RAs in GPC leads to reduction of space required for landfill, and also it conserves the natural resources. Modifications in the mix design are required to attain high strength durable GPC cured at ambient condition. There is a need for the development of low cost alkaline materials to make GPC as an economically viable and cost effective material.

3. Both curing time and curing temperature influence the compressive strength of GPC. The complete geopolymerization process can be made to achieve the high strength durable GPC by adopting curing at elevated temperatures compared to ambient temperature. Longer the curing time improves the polymerization process resulting in higher compressive strength. The elevated temperatures cause the implementation of GPC technology uneconomical.

4. Even though GPC has superior properties as compared to OPC concrete, the practical application of GPC technology, have some constraints such as the health hazards due to handling of source materials, alkaline liquids and curing at elevated temperatures. In near future, further research is needed to rectify these shortcomings and to make GPC fit for the practical applications in construction sector.

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