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Enhancing the Durability of Concrete in Sulfate-Rich Environments: A Review of Waste Materials Incorporation

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Abstract:- Concrete, the most widely used construction material, confronts durability challenges in aggressive sulfate-rich environments. The reaction between sulfate ions and cementitious materials causes various types of deterioration, such as expansion, cracking, and loss of strength. To address these concerns and promote sustainability, researchers have turned to incorporating mineral waste materials like fly ash and slag in concrete mixes. These by-products not only mitigate environmental impact but also enhance sulfate resistance. This review collates key studies on concrete durability in sulfate-rich environments, with a focus on mechanism of sulfate attacks. Understanding these mechanisms is pivotal in designing sulfate-resistant concrete mixes and ensuring long-term performance in such conditions. The knowledge derived from this review paves the way for resilient and sustainable concrete solutions in sulfate-exposed environments.

***Keywords**:- Concrete, Sulfate Attack, Durability, Mineral Waste, Aggressive Environment.*

I. INTRODUCTION

Concrete, as the world's most widely used construction material, plays a vital role in infrastructure development. However, its durability can be compromised when exposed to aggressive environments containing sulfates, which are commonly found in soils, groundwater, and industrial effluents, and marine environments (Qiao *et al.*, 2012; Albitar *et al.*, 2017). The interaction between sulfate ions and the cementitious materials in concrete can lead to various forms of deterioration, including expansion, cracking, strength loss, and surface degradation (Cang, Ge and Bao, 2017; Zhao *et al.*, 2019).

To enhance the durability of concrete in sulfate-rich environments and explore more sustainable alternatives, researchers have increasingly focused on the incorporation of mineral waste materials (Tang *et al.*, 2019; Fang, Zhan and Poon, 2021). Mineral wastes, such as fly ash, slag, and silica fume, rice husk ash snail shell ash, are by-products of industrial processes, and their utilization in concrete not only reduces environmental impact but can also improve the concrete's resistance to sulfate attack (Adewuyi, Franklin and Ibrahim, 2015; Elahi *et al.*, 2021).

The study and review of the durability of concrete and mineral waste concrete in sulfate-rich environments are of paramount importance to understand the mechanisms of sulfate attack and develop effective strategies to mitigate its adverse effects. Numerous studies have been conducted over the years to investigate the behavior of concrete in sulfate exposure conditions and evaluate the performance of mineral waste concrete (Jun et al., 2019; Stark, 2002; Wuman et al., 2019; Zhang et al., 2019; Zhao et al., 2012).

This review aims to collate and synthesize the findings from various key studies on the durability of concrete and mineral waste concrete in sulfate-rich environments.

➢ *Mechanism of Sulfate Attacks in Concrete Exposed to Sulfate Rich Environment*

Sulfate attack in concrete can occur through three primary mechanisms, as reported in various research studies.

According to (El-Hachem *et al.*, 2012; Gu *et al.*, 2019, 2022) the external sulfate attack, also referred to as surface sulfate attack, involves the penetration of sulfate ions from the external environment into the concrete through capillary action or diffusion, where sulfate-rich solutions or soils reacted with calcium aluminates' compounds in the cement paste, forming ettringite and gypsum. The ensuing volumetric expansion led to internal stresses, eventually causing cracking and surface deterioration (Diaz Caselles *et al.*, 2021).

Internal sulfate attack, known as delayed ettringite formation (DEF), is a phenomenon observed within the concrete itself. According to a study by (Paul *et al.*, 2022), DEF is characterized by the delayed formation of expansive ettringite crystals, primarily caused by the presence of reactive aggregates or high cement content. This leads to a slow reaction between sulfate ions and calcium aluminates' compounds, resulting in the gradual growth of ettringite. Consequently, the concrete experiences internal stresses, leading to cracking and a loss of concrete strength. DEF's detrimental effects on concrete's mechanical properties and durability make it a critical concern in sulfate-rich environments. This deterioration that occurs due to the delayed ettringite formation is when calcium aluminates hydrate (C-A-H) in concrete reacts with aggressive water solutions containing sulfate ions, which as demonstrated in the following reaction (Ciliberto, Ioppolo and Manuella, 2008).

$$3\ CaO \cdot Al_2O_3 + 3\ CaSO_4 + 26\ H_2O \rightarrow 3\ CaO \cdot Al_2O_3 \cdot 3\ CaSO_4 \cdot 32\ H_2O \qquad (1)$$

Thaumasite sulfate attack is a severe form of sulfate attack that occurs in the presence of high levels of sulfate ions and calcium carbonate ($CaCO_3$). It leads to the formation of thaumasite, a detrimental compound, within the concrete, causing extensive damage and weakening of the cementitious matrix. This attack can severely deteriorate the structural integrity of the concrete (Zhang, Wen, and Niu, 2021). Through the reaction as also cited in (Ciliberto, Ioppolo and Manuella, 2008), thaumasite forms through a low-temperature process when calcium silicate hydrate gel reacts with water solutions containing sulfate and carbonate ions.

$$3CaO \cdot 2SiO_2 \cdot 3H_2O + H_2O + SO_4^{2-} + CO_3^{2-} \rightarrow [Ca_3S_i(OH)_6].(SO_4)(SO_3).26H_2O \qquad (2)$$

Ettringite and thaumasite formation from sulfate attack of concrete has been studied in several papers. Gao *et al.*, (2021) Found that the content of limestone powder in cement-based materials affects the degree of Thaumasite form of sulfate attack (TSA). Xu *et al.*, (2020) investigated the occurrence time of TSA on tunnel concrete structures and reported that thaumasite was formed 18 to 36 months after construction. Chinchón-Payá *et al.*, (2020) analyzed the relation between ettringite and Thaumasite formation and found that thaumasite forms from ettringite by substitution of aluminum with silicon. Gu *et al.*, (2019) studied the degraded state of cement paste exposed to external sulfate attack (ESA) and delayed ettringite formation (DEF) and proposed a global expansion mechanism. Nguyen *et al.*, (2020) developed an accelerated method to study delayed ettringite formation using electrochemical techniques.

II. LITERATURE REVIEW ON THE DURABILITY OF CONCRETE IN AN AGGRESSIVE ENVIRONMENT OF SULFATES

Wu *et al.*, (2021) investigated the durability of concrete materials when exposed to sulfate attack in a sulfate-rich environment. Through a series of concrete tests, the study examined the mechanical properties and microstructure of the concrete, as well as the process of damage evolution under sulfate attack. The study findings indicate that, under the influence of sulfate attack, the rate of mass change, relative dynamic modulus of elasticity, corrosion resistance coefficient of compressive strength, and corrosion resistance coefficient of splitting tensile strength initially increase and then decrease. Concrete mixed with 10% fly ash content exhibited the highest resistance to sulfate attack within the same erosion time. The researchers also developed a damage evolution equation for sulfate attack, revealing an exponential function relationship among different damage variables. Additionally, the study highlighted the significant binary curved surface regression effect of concrete damage concerning erosion time and the amount of fly ash, enabling the prediction of concrete deterioration under sulfate attack.

Yang *et al.*, (2021) investigated how treating crumb rubber with NaOH solution affected the sulfate attack resistance of recycled aggregate concrete. Their study demonstrated that incorporating crumb rubber particles improved the concrete's ability to resist sulfate attack. The researchers concluded that adding NaOH-solution-treated crumb rubber enhanced the sulfate attack resistance of recycled aggregate concrete, with a more significant effect observed for treated particles compared to untreated ones. Optimal conditions for enhancing resistance were determined to be using 0.16-0.3 mm crumb rubber particles and pretreating them with a 20% NaOH solution. The macroscopic performance of recycled aggregate concrete with crumb rubber exhibited three stages: an initial stage of enhancement or slow decline, followed by a middle declining stage, and finally an accelerated decline stage. Notably, mixtures containing 10-20 mm crumb rubber particles remained durable in sulfate attack conditions due to their higher residual rapid chloride migration (RDME) and compressive strength compared to regular recycled aggregate concrete. The findings from the X-ray diffraction analysis of corrosion products and the scanning electron microscopy examination of the microstructure were consistent with the observed macroscopic performance.

Elahi *et al.*, (2021) carried out a comprehensive review on enhancing the sulfate attack resistance of concrete through the utilization of supplementary cementitious materials (SCMs). The authors explored various studies and findings related to this topic. The review highlights the potential of SCMs in improving concrete's resistance to sulfate attack. The researchers emphasize that incorporating SCMs in concrete mixtures can enhance sulfate attack resistance by reducing the permeability of the concrete, increasing its strength, and providing better long-term durability. The review paper provides valuable insights into the mechanisms and effects of SCMs on sulfate attack resistance, offering guidance for engineers and researchers in optimizing concrete mix designs for improved performance in sulfate-rich environments.

Cuesta *et al.*, (2020) conducted a study on the utilization of Recycled Concrete Aggregate (RCA) in the production of Self-Compacting Concrete (SCC). The research involved completely replacing the coarse fraction of the natural aggregate with RCA (100% replacement rate) and replacing half of the fine fraction (50% replacement rate). The durability of the recycled SCC was assessed against aggressive external factors, and the findings revealed its resistance to freeze/thaw and moist/dry cycles, as well as sulfate attack. These results indicate that the recycled SCC is suitable for structural concrete components. The study emphasizes that while the mechanical performance of vibrated concrete with RCA is known to decrease compressive strength and durability, incorporating RCA in SCC can actually improve its durability and enhance other properties.

Hendi *et al.,* (2020) conducted a study to evaluate the performance of two types of concrete, Ordinary Concrete (OC) and Self-Consolidating Concrete (SCC), incorporating waste silica sources under severe magnesium sulfate attack. The findings revealed that SCC mixtures with a six percent substitution of GP (glass powder) and MS (micro silica) exhibited the highest resistance to deterioration among all the mixtures studied. The addition of micro silica and glass powder as supplementary waste materials had a significantly positive impact on the durability of Ordinary Concrete (OC) when exposed to magnesium sulfate attack. However, these additions did not improve the durability performance of self-consolidating concrete. Overall, the self-consolidating concrete performed better than normal concrete up to 88% due to the contribution of superplasticizer. The durability index analysis further confirmed that SCC mixtures with a six percent GP and MS substitution showed the best resistance to deterioration among all the mixtures evaluated.

Mostofinejad *et al.,* (2020) investigated the effects of recycled coarse aggregate (RCA), recycled fine aggregate (RFA), and milled waste glass (MWG) on the durability of concrete. The researchers evaluated the durability of concrete mixes by measuring expansion, mass variation, water absorption, and compressive strength of the samples. The findings suggest that incorporating RFA in concrete mixes can improve the durability of concrete in sulfate environments. Furthermore, the combination of MWG with RCA and RFA can enhance the sulfate durability of concrete, particularly in terms of compressive strength. The study also introduces the Durability Loss Index (DLI) as a means to assess the durability of concrete in different groups. However, it is important to note that the study has limitations, including its focus on sulfate environments only and the use of a single type of recycled aggregate.

Baghabra *et al.,* (2019) conducted a study to assess the performance of concrete mixtures containing natural pozzolans from various sources in Saudi Arabia. The research findings indicate that the concrete mixtures incorporating natural pozzolans displayed improved durability characteristics compared to the control mixture consisting solely of ordinary Portland cement (OPC). These improvements were observed in terms of reduced water penetration depth, lower chloride diffusion coefficient, enhanced resistance to reinforcement corrosion, and increased protection against sulfate attack. Consequently, the study concludes that incorporating natural pozzolans in concrete mixtures can enhance the durability of concrete subjected to chloride and sulfate exposure. However, it is worth noting that the compressive strength of concrete mixtures containing natural pozzolans was slightly lower compared to those containing OPC alone.

Wuman *et al.,* (2019) carried out investigation on the impact of low-calcium fly ash (LCFA) on the resistance of cement paste against sulfate in various exposure conditions. The study focused on analyzing the physical properties of cement/LCFA paste, as well as the compressive strength and microstructures of the specimens. The findings of the study indicate that the inclusion of low-calcium fly ash (LCFA) can enhance the sulfate resistance of cement paste when exposed to different conditions. It was observed that the addition of LCFA led to increased water demand, setting time, and soundness of the cement paste samples. Moreover, the introduction of LCFA resulted in a decrease in the strength reduction of cement specimens when immersed in a 3% Na2SO4 solution. The compressive strength of all specimens incorporating LCFA exhibited an increase with longer immersion time. The microstructures of the cement/LCFA specimens were found to correlate well with the observed compressive strength. Overall, these research findings hold potential significance in the development of more sustainable and durable concrete materials.

Kępniak *et al.,* (2019) aimed to assess the suitability of waste limestone powder as a replacement for fine aggregate in cement composites, and its effect on the durability of concrete in terms of sulfate degradation and chloride ion diffusion. The findings indicate that the tested waste limestone dust can be utilized as a substitute for a portion of the natural fine aggregate without causing any detrimental effects on the durability of the concrete. The study concludes that waste limestone powder can effectively replace fine aggregate in cement composites without compromising the durability of the concrete. The investigation into sulfate degradation and chloride ion diffusion further supports this conclusion, demonstrating that the tested waste limestone dust does not negatively impact the durability of concrete. The overall desirability function was utilized, and the majority of the investigated concretes achieved satisfactory desirability values.

Xie *et al.,* (2019) aimed to develop an environmentally friendly concrete with enhanced sulfate resistance by utilizing recycled aggregates from construction and demolition waste as coarse aggregates. In addition, granulated blast furnace slag (GGBS) and fly ash-based geopolymer were used to replace cement in the concrete mix. The study specifically focused on evaluating the sulfate resistance of this geopolymer recycled aggregate concrete (GRAC). The results of the study demonstrate that the proposed GRACs exhibited excellent sulfate resistance and can be effectively utilized in construction projects situated in sulfate-rich environments.

The study concludes that the geopolymer recycled aggregate concrete (GRAC) with a higher proportion of GGBS showcased reduced mass loss and higher residual compressive strength following exposure to sulfate, indicating outstanding sulfate resistance. By employing GRACs in construction projects in sulfate-rich environments, the reliance on cement can be reduced, and the disposal of construction and demolition waste can be effectively managed.

According to the study conducted by (Alnahhal *et al.*, 2018), the industrial by-products were examined as substitutes for traditional supplementary cementitious materials (SCMs) in recycled aggregate concrete. By incorporating rice husk ash, palm oil fuel ash, and palm oil clinker powder, up to 30% of the mixture, researchers observed minimal concrete deterioration and maintained compressive strength when exposed to hydrochloric acid and magnesium sulfate attacks. The conclusion, according to the study, highlights the efficacy of these alternative materials in enhancing concrete's resistance to acid and sulfate attacks, making them environmentally sustainable substitutes for traditional SCMs, and promoting eco-friendly construction practices.

Dave et al., (2017) investigated the mechanical behavior and durability of quaternary binders made by blending supplementary cementitious materials with Ordinary Portland Cement (OPC). Different materials (Fly ash, GGBS, MK, and SF) replaced 30-50% of OPC. Mechanical behavior was tested over time and compared to controlled concrete and binary mixes. Durability was assessed using RCPT and sulfate attack test. Results indicate that incorporating these materials enhances concrete durability and strength, with some combinations surpassing 100% OPC. The synergy between cement and supplementary materials improves durability. RCPT and sulfate attack test demonstrated superior durability of quaternary mix compared to controlled and binary mixes. Utilizing quaternary binders with OPC and supplementary materials is preferred over 100% controlled concrete.

Cang et al., (2017) examined mechanical properties and damage evolution of high-performance concrete (HPC) with varying fly ash contents in a magnesium sulfate environment. Results reveal that HPC with 20% fly ash exhibited the highest resistance to magnesium sulfate attack. The average porosity increase for HPCs before and after exposure to the sulfate solution was 34.01%. Notably, HPC containing 20% fly ash demonstrated superior resistance to magnesium sulfate attack in terms of porosity.

Palankar et al., (2016) investigated the durability performance of eco-friendly concrete mixes utilizing steel slag as coarse aggregates, comparing them with conventional Ordinary Portland Cement Concrete (OPCC). The study concluded that the eco-friendly mixes exhibited superior durability and lower production costs. Incorporating steel slag as coarse aggregates in these mixes provided sufficient strength for structural and highway applications, although there was a slight reduction in strength properties compared to OPCC. The researchers conducted a detailed analysis of durability properties, including long-term aging performance, water absorption, volume of permeable voids, resistance to sulfuric acid attack, and resistance to magnesium sulfate attack, comparing the results with OPCC. Additionally, the paper included an ecological and economical analysis of the concrete mixes, confirming their improved durability and cost-effectiveness when utilizing steel slag as coarse aggregates compared to traditional OPCC.

Merida et al., (2014) investigated the positive impact of incorporating natural pozzolan as a partial replacement for cement in high-performance concrete exposed to sulfated environments. The study focused on key parameters to provide valuable insights into the enhanced durability achieved through the use of natural pozzolan. Experiments on high-performance concrete with natural pozzolan as a partial cement replacement in a sulfated environment showed that the addition of pozzolan positively influenced the mechanical, physical, and physico-chemical properties of the concrete, enhancing its durability. Specifically, replacing 5% of cement with natural pozzolan led to improved durability in sulphate environments. The presence of pozzolan in the concrete modified its microstructure, reducing porosity and increasing densification, thereby enhancing the physical characteristics of the concrete compared to the control group. Overall, the study emphasized that incorporating natural pozzolan in high-performance concrete contributes to its improved durability in sulphated environments, with its positive effects on the concrete's mechanical and physical properties highlighting its potential to enhance overall concrete performance.

Qiao et al., (2012) aimed to assess the durability of concrete in highly saline regions by investigating the performance of different high-performance fine aggregate concretes in a sulfate environment. The study provides recommendations for concrete mixing in salt-affected areas based on the research findings. The study revealed that sulfate-resistant cement concrete performs similarly to Portland cement concrete under aggressive dry-wet cycles. However, high-performance concrete incorporating a composite additive shows improved sulfate resistance. This additive promotes secondary hydration, reducing $Ca(OH)_2$ content, and enhancing the microstructure of the concrete. Based on these findings, the paper suggests that in regions with high salt content, using high-performance concrete with the composite additive can enhance the durability of concrete structures. The additive's ability to promote secondary hydration and improve the microstructure contributes to increased sulfate resistance, providing valuable guidance for concrete mixing practices in salt-affected areas.

III. CONCLUSION

In summary, this review emphasizes the crucial significance of addressing concrete durability in environments rich in sulfates, as it presents significant challenges to the long-term performance of infrastructure. The interaction of sulfate ions with concrete can lead to various forms of deterioration, including expansion, cracking, and strength loss, necessitating effective solutions. To combat this issue, researchers have explored the incorporation of mineral waste materials like fly ash, slag, and silica fume, which not only improves concrete's resistance to sulfate attack but also reduces environmental impact. Furthermore, alternative materials such as rice husk ash, and palm oil fuel ash have shown promise in enhancing concrete durability in sulfate-rich conditions. The review also highlights the effectiveness of geopolymer recycled aggregate concrete and quaternary binders in providing exceptional sulfate resistance while promoting environmentally friendly construction practices.

Understanding the three primary mechanisms of sulfate attack (external, internal, and thaumasite sulfate attack) is crucial for designing sulfate-resistant concrete mixes and implementing preventive measures. The research findings underscore the importance of concrete mix design and the selection of suitable supplementary cementitious materials tailored to specific sulfate exposure conditions. By incorporating natural pozzolans and steel slag, concrete durability can be enhanced without compromising overall strength. This comprehensive review provides valuable insights and guidance for engineers and researchers in devising more sustainable and durable concrete structures capable of effectively withstanding the challenges of sulfate-rich environments. Ultimately, the knowledge gained from this study contributes to the advancement of construction practices and the development of resilient infrastructure.

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COMPLIANCE WITH ETHICAL STANDARDS
CONFLICT OF INTEREST

The authors declare that they have no conflict of interest.

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