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A comparative study of jarosite and other cementitious materials as a concrete material – A review

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Abstract

In recent years, zinc production has grown exponentially, compensating for the need arising from worldwide industrial growth. Jarosite is a non-biodegradable mineral residue obtained from smelting zinc ore. Jarosite is categorised as a hazardous material due to its high metal ion concentration, and hence, the safe disposal of massive volumes of jarosite waste poses a significant burden. This review article highlights how various proportions of jarosite affect concrete's fresh, mechanical, hydration, microstructural, and durability properties. When used in specific volumes, jarosite positively impacts the concrete compressive strength. The hydration mechanism of jarosite-incorporated concrete justifies forming a secondary hydration reaction, which causes dense morphology observed during microstructure analysis. Further, when jarosite is incorporated into concrete, the leaching characteristics of raw jarosite are observed to be substantially reduced and meet the safe limits. Based on the observations, a jarosite replacement level of 15% to the cementitious mix shows improvement in the mechanical and durability properties of the concrete. These findings could be used to identify a sustainable approach to reutilise the zinc industry waste by-products (jarosite). The experimental values of jarosite-incorporated concrete were compared with those of similar well-researched industrial waste materials such as red mud and copper slag to ensure that the observation and evaluation are accurate and trustworthy.

Keywords: Jarosite, Sustainable waste management, Industrial waste utilization, Concrete material

1. Introduction

Jarosite is a non-biodegradable by-product from the zinc industry [1-3], it is obtained when zinc ore is leached with sulphuric acid during the smelting stage of the hydrometallurgical process [4-6]. It is observed that for every tonne of zinc refined, nearly half a tonne of jarosite is obtained as a by-product [7]. According to the reports submitted by the United States Government Survey (UGCS), India is one of the world's top four largest zinc producers, with a total of 8.3 lakh tonnes of zinc production in the year 2022 [8]. Figure 1 shows the worldwide production of zinc in the year 2022. The enormous production of jarosite creates a hazardous environment due to the presence of a high concentration of heavy metal ion content. This causes worldwide concern [9], mainly in countries like India and China, where most of the waste jarosite is disposed of at landfills inside the industrial premises [10]. The disposal concerns of similar industrial by-products, such as red mud [11], magnesite mine waste [12], and copper slag [13], are solved by determining a sustainable application where the waste materials can be reutilised.

Concrete is the most prevalent construction material used worldwide [14, 15]. The primary binding material for concrete is cement, and the manufacturing and usage of cement have a considerable contribution to the increase in greenhouse gas content [16]. In the past decade, more significance has been given to decreasing environmental pollution by reducing the usage of significant contributors of greenhouse gases like fossil fuels and cement [17, 18]. The research contributing to the reduction of cement in concrete has increased in recent years, aiding the development of green concrete [19-22]. The identification of suitable alternative materials for conventional concrete materials has been the major challenge in the concept of sustainable development. From the viewpoint of conserving natural resources and reducing CO_2 emissions, the incorporation of industrial waste as concrete material is the most economical and sustainable approach. The construction industry is by far the most waste-consuming industry when compared to other sectors, making it a more suitable industry for waste material recycling. Various concrete properties like compressive strength, slump value, and durability parameters can be improved based on the physical and chemical properties of waste material that is used as an alternative to conventional concrete materials. Waste material like ground granulated blast furnace slag(GGBS) [23-25], Fly ash (FA) [26, 27], Red mud(RM) [28, 29], Copper slag(CS) [13, 30], steel slag(SS) [31, 32], Agricultural waste(AG) [33, 34] have been extensively researched and impacts of utilising these waste materials in concrete have proven to be advantageous compared to conventional concrete materials. This present paper discusses the potential of utilising zinc industrial waste (jarosite) as a concrete material.

In the last decade, there has been an increase in research on jarosite as a construction material. Kumar et al. [35] published a comparative analysis of fine aggregate-replaced concrete using various industrial wastes, including crumb rubber, jarosite, and quartz sandstone. The mechanical properties of the jarosite-incorporated concrete mixes show an increase in strength with increasing jarosite replacement levels at various water-cement (w/c) ratios [35]. Mehra et al. [36-38] conducted a study with similar jarosite replacement

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levels for more extended curing periods of 180 and 365 days. Their findings revealed that the mechanical and durability characteristics of the concrete mix improved with the addition of jarosite as a substitute for fine aggregates [36-38]. Both Kumar et al. [35] and Mehra et al. [36] demonstrated similar results of enhanced mechanical performance, thus acknowledging jarosite's potential as a substitute for fine aggregate in the concrete mix. Gared and Gaur [39] reported a maximum increase in compressive strength observed with 25% jarosite substituted as the binding material. The flexural and tensile strength data demonstrated a similar increasing pattern with increasing jarosite replacement levels [39]. Ray et al. [40] conducted a study on jarosite as a replacement of binding materials such as ordinary Portland Cement (OPC) and Portland Pozzolana Cement (PPC) and achieved improved mechanical properties with about 15% replacement of the binding material with jarosite. Research by Saini et al. [41, 42] confirmed that the strength parameters of jarosite-incorporated concrete increased for 15% replacement levels of jarosite as the binding material. The study was done by Gupta and Sachdeva [43-45] and Debbarma et al. [46] only mechanical characteristics such as compressive and flexural strengths were reported, which dropped when the increased proportion of jarosite as an alternative binding material was incorporated into concrete. Nevertheless, the drop in strength was relatively minimal and hence insignificant in the utilisation potential. In contrast, a steady increase in the strength parameters was reported by [39-42].

There are various research involving jarosite other than concrete material. Mymrin et al. [47] conducted research where jarosite in its stabilised form is used as a base/sub-base course in road construction, Asokan et al. [48] developed a jarosite and clay blended bricks where the mechanical properties of the brick improved at higher jarosite content. Katsioti et al. [49] substituted jarosite/ alunite mixture for gypsum in the cement manufacturing process. A comparative analysis of the properties of jarosite with that of similar cementitious materials is presented in this review article, wherein the chemical, mechanical, durability, and microstructural properties of jarosite-incorporated concrete investigated by various researchers have been presented and thoroughly analysed to justify the potential of jarosite a viable concrete material.

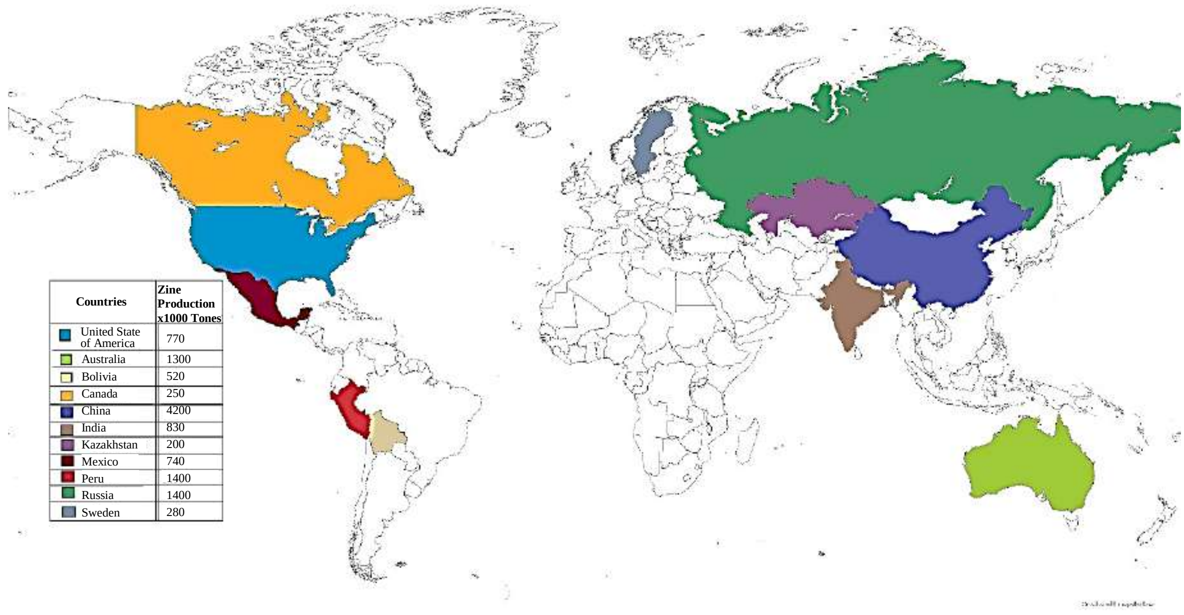


Figure 1 Map indicating the Country's production of zinc [8].

2. Methodology

This review article presents a comparative study of experimental analysis on concrete incorporating jarosite and similar industrial waste materials such as red mud, copper slag, fly ash, silica fume, and ground granulated blast furnace slag. Jarosite has been identified as a construction material only in the past decade therefore, a relatively small amount of research has been done on the characteristics of jarosite-incorporated concrete compared to the other well-researched materials. Articles available in the previous literature investigated the application of jarosite in various fields, mainly as construction material and geotechnical applicability. This paper summarises previous studies investigating the use of jarosite as a substitute material in concrete and the implications of jarosite incorporation on the concrete's mechanical, durability, and microstructural properties. The research findings were analysed to determine the extent to which jarosite is used as a binder replacement in cementitious materials, providing researchers with a vivid and distinct view of this sustainable alternative cementitious material in the concrete industry.

3. Process of obtaining jarosite from the zinc smelting process

Zinc can be extracted from its ore through two different processes, namely, roast leaching electrolysis and imperial smelting technique, where the roast leaching technique is widely employed globally [50]. Jarosite is obtained as a waste by-product during the roast leaching electrolysis process of the zinc ore. Figure 2 depicts the simple two-stage Jarosite precipitation process. Jarosite is formed when the zinc ore is roasted at 900℃ and then leached using a hot acid. Jarosite has the general formula $XFe_3(SO_4)_2(OH)_6$ [50] with K (potassium), Na (sodium), NH_4^+ (ammonium), including ions like Ag^+, Pb^{2+}, Ti and oxides such as Pb, Zn, SiO_2 and ferric oxyhydroxide (FeO(OH)) also being present in the jarosite waste [47]. It has a pH value of 2.7 to 3.6, making this material highly acidic in nature. Jarosite and other zinc waste are categorised as hazardous waste per the Basel Convention. In general, all wastes that contain zinc have been classified as hazardous by the European Union [51] due to the level of contamination of the environment. Since jarosite gets dumped directly on the landfill, it pollutes the underlying water table, affecting the aquatic life of the surrounding water bodies.

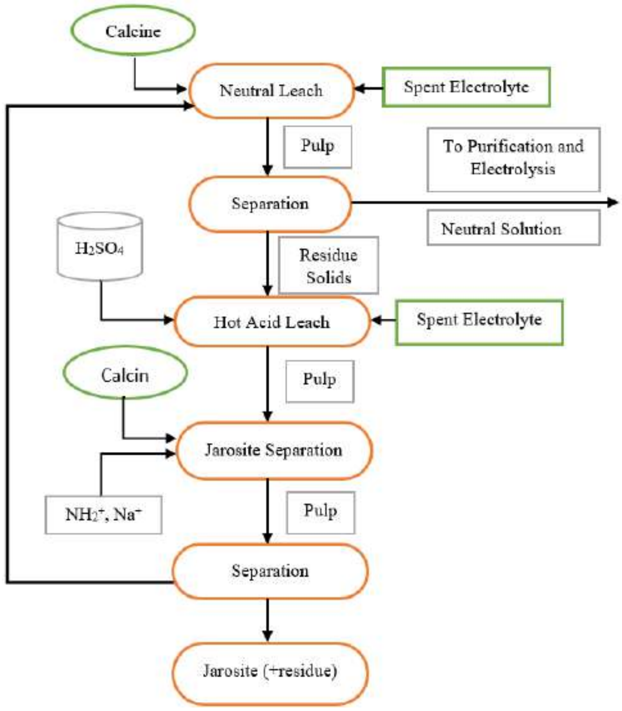


Figure 2 Flow chart showing the simple two-stage Jarosite precipitation [52].

3.1 Treatment of hazardous jarosite

Given its severe toxicity, jarosite is blended with 2% lime and 10% cement to create jarofix, which is a less toxic material produced due to chemical stabilisation. Jarofix deposited in specially constructed ponds encased with low-density polyethylene (LDPE) sheets [53]. The amount of dumped jarofix continues to rise rapidly, inhabiting valuable agricultural lands and urban areas without significant utility. Seyer et al. [54] mixed jarosite with Portland cement, water, and lime to obtain a chemically and physically stable material and discovered that jarosite combines with the alkaline components in cement to generate a variety of stable phases containing zinc and other soluble metals, ensuring its long-term environmental stability. Arroyo et al. [55] proposed a method for stabilising jarofix by incorporating sludge (waste), thus making it more sustainable and available for multiple applications, such as pavement sub-base and manufactured bricks. Jarosite can also be employed as a proxy in remote-sensing studies to identify areas with elevated acidity and metal leachability, providing an efficient method to screen mined areas for potential sources of acidic drainage, as reported in [56].

4. Characteristics of jarosite

Knowing the physical properties of jarosite, such as grain size, specific gravity, and surface area, helps to ascertain its suitability to be incorporated as a concrete material. Jarosite is a fine-grained substance comprising 20% clay, 75% silt, and 5% sand particles. It has been identified to have a non-swelling nature. This section presents the chemical composition and physical parameters of jarosite

4.1 Physical parameters

In almost all the jarosite-based research conducted in India, the waste material is sourced from Hindustan Zinc Ltd (HZL). HZL is India's primary zinc manufacturer. As shown in Figure 3, the jarosite is yellow in colour [57, 58]. The microstructure of the jarosite shows rectangular-shaped, densely packed particles [57]. Jarosite can be described as a silty, fine-grained material with excellent plasticity in accordance with ASTMD2487-11 soil classification [59]. Jarosite comprises more than 99% of fines, and hence, a hydrometer investigation was used to identify particles of size below 75 microns [60].

(a) (b)

Figure 3 (a) Raw Jarosite (b) SEM image of raw jarosite.

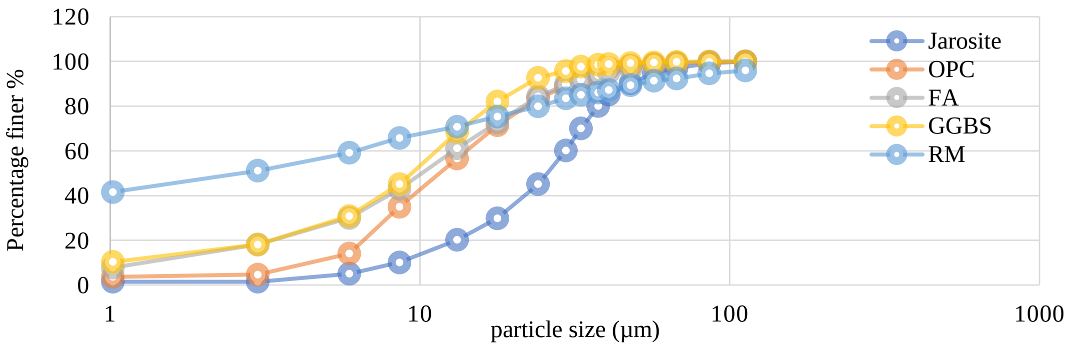


Figure 4 Comparison of the particle size distribution of Jarosite [60], GGBS [61], fly ash [61], red mud [62], and OPC [63].

Compared with other cementitious materials and industrial waste materials, jarosite has the finest particle size, followed by GGBS [64], [26] fly ash, red mud, and Ordinary Portland Cement (OPC), as presented in Figure 4. Table 1 presents the physical properties of different industrial waste materials, which are compared with the physical properties of jarosite represented in Figure 5. Specific gravity is a crucial characteristic of any cementitious material owing to its relationship to the concrete's density and viscosity. The specific gravity of jarosite varies from 2.7 to 2.9, which is slightly less than that of the conventional OPC, resulting in a higher volume of utilisation, as seen in Figure 5(a). In addition, jarosite also possesses a higher surface area, as depicted in Figure 5(b). For a material finer than cement, jarosite surrounds the cement particles during the initial stages [40] owing to its high surface area. This causes a prolonged water-cement reaction, leading to the secondary hydration process.

Table 1 Physical properties of materials used as a replacement in concrete

Material	Specific Gravity	Surface Area (m^2/g)	Density (g/cm^3)	Reference
OPC	3.12	-	-	[20]
GGBS	2.84	-	-	[25]
GGBS	2.85	-	-	[27]
FA	-	5.35	2.25	[62]
RM	-	9.79	3.18	[62]
FA	2.41	-	-	[65]
FA	2.13	-	-	[66]
OPC	-	0.314	3.15	[67]
FA	-	-	2.13	[67]
GGBS	-	0.4254	2.91	[67]
GGBS	2.88	-	-	[68]
RM	2.95	-	-	[68]
OPC	-	0.3144	3.15	[69]
RM	-	23.53	3.5	[69]
GGBS	-	4.21	2.67	[70]
RM	-	8.72	2.89	[70]
GGBS	-	4.245	2.931	[71]
RM	-	10.854	2.781	[71]
OPC	-	3.264	3.101	[71]
RM	-	8.04	-	[72]
GGBS	-	1.44	-	[72]
FA	2.85	-	-	[73]
SF	-	23	-	[74]

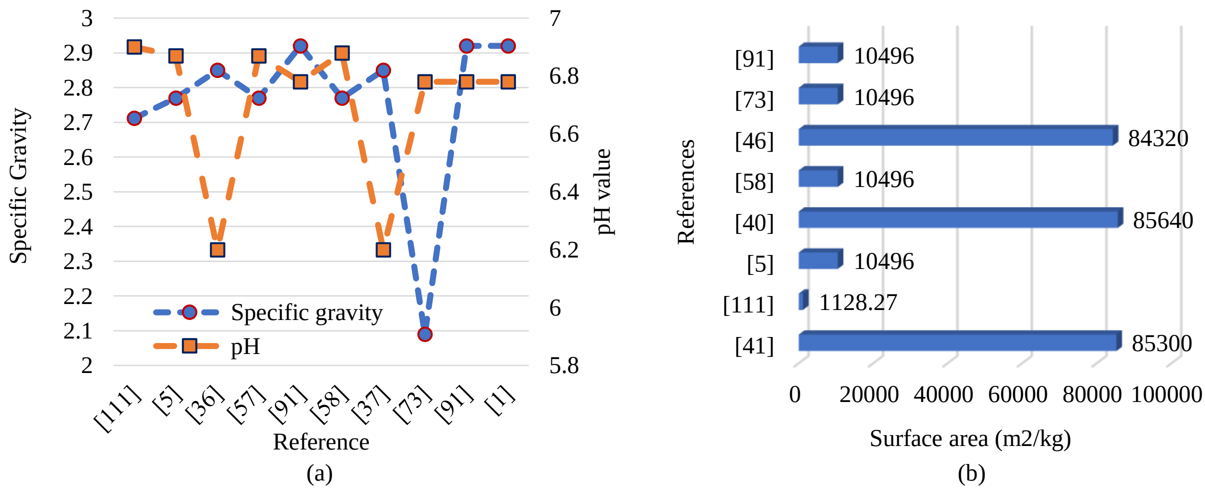


Figure 5 Physical properties of jarosite (a) specific gravity and pH, (b) surface area.

4.2 Chemical composition

The primary components of jarosite include iron oxide (Fe_2O_3), Silicon dioxide (SiO_2), and Sulphur trioxide (SO_3), as evident in Table 2. Figure 6 represents the chemical compositions of jarosite and of similar metal industry by-products such as Red Mud (RM) [28, 75-77], Copper Slag (CS) [78-80], and other supplementary cementitious materials. Jarosite possesses large proportions of SO_3 and Zinc oxide (ZnO) components. Generally, increasing the sulphur trioxide content in the cement increases its setting time, accompanied by a rapid drop in the compressive strength [81]. According to ASTM [82], a material is classified as pozzolanic if the principal oxide constituents such as Fe_2O_3, Aluminium oxide (Al_2O_3), and SiO_2 comprise at least 50% of the total concentration. The proportion of the primary oxide constituents of jarosite ranges from 45.62 to 53.77%, with iron oxide being the principal constituent. The primary oxide constituents of red mud (RM) and copper slag (CS) vary from 60-77.6% and 69.87-90.5%, respectively. In RM, all three primary oxides are present in equal concentration, while CS has a lower alumina concentration. Despite the low primary oxide content and additional SO_3 content, most of the jarosite-incorporated concrete exhibited enhanced mechanical and durability properties. Therefore, jarosite can be categorised as a pozzolanic material based on the primary oxide concentration.

Table 2 Chemical composition of Jarosite (JS).

Ref.	Material	Al_2O_3	CaO	Fe_2O_3	K_2O	MgO	Na_2O	SiO_2	SO_3	ZnO
[38]	Jarosite	3.2	2.8	37.55	0.625	0.302	0.559	13.02	29.35	3.68
[40]	Jarosite	2.93	2.59	38.55	-	0.126	4.919	10.06	33.74	3.03
[57]	Jarosite	6.75	6.87	32.12	0.74	1.86	0.63	6.75	31.8	9.18
[58]	Jarosite	8.98	4.92	32.11	0.76	1.08	0.63	7.73	31.8	9.07
[83]	Jarosite	1.78	2.79	22.41	0.6	0.58	0.15	20.27	32.53	3.64

The primary difference between the chemical compositions of jarosite and that of other cementitious materials such as OPC [84], fly ash [26], silica fume [85], metakaolin [86], and GGBS [87] is the increased dosage of Fe_2O_3 and SO_3 as observed in Figure 6. It is evident that compared to other cementitious materials, jarosite [42] has low concentrations of calcium oxide and silicon dioxide, which are vital for forming Calcium Silicate Hydrate (C-S-H) gel. Also, the combination of GGBS and silica fume provides a chemical composition similar to that of OPC [84]. Ettringite generation from Tricalcium aluminate (C_3A) requires a more significant amount of water than other calcium aluminate hydrates, notably C_2AH, C_4AH_{13}, and C_3AH_6 formed with lower SO_3 content [42]. Zinc and Lead ions are known to be amphoteric, and the re-dissolution of excess hydroxide ions would lead to the development of soluble zincates and plumbates from the hydroxide precipitates [88]. This process depletes the calcium and hydroxide ions solution and postpones the supersaturation, precipitation, and production of $Ca(OH)_2$ and C-S-H.

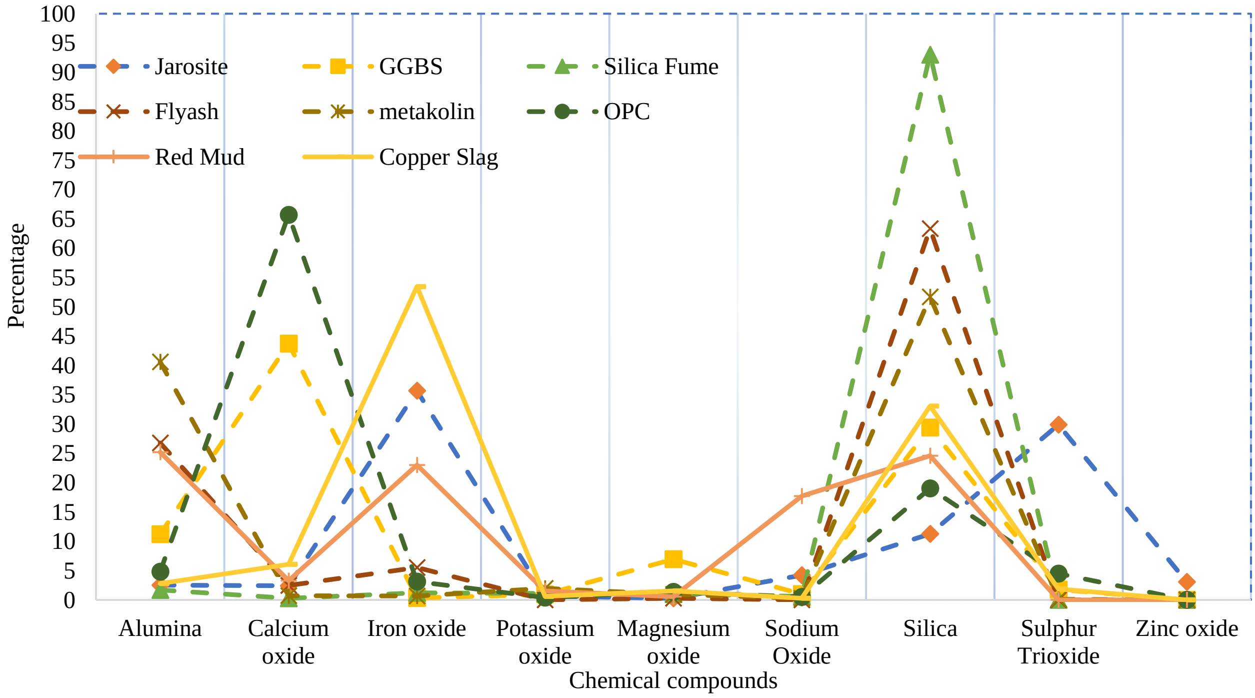


Figure 6 Chemical composition of the JS compared with GGBS, SF, FA, and MK.

5. Application aspect of jarosite as a construction material

5.1 As a geotechnical material

Gupta and Prasad [58] determined the Maximum Dry Density (MDD) and Optimum Moisture Content (OMC) in addition to the strength and durability characteristics, including unconfined compression, split tensile, and freeze and thaw cycle of jarosite stabilised using ground granulated blast furnace slag (GGBS). GGBS is a chemical stabiliser that neutralises the leaching produced by raw jarosite. The presence of GGBS modifies the compaction of jarosite waste. The OMC of the mixture declined while the MDD increased as the GGBS percentage increased to 30%. Adding GGBS beyond 30% caused the MDD to decrease and OMC to increase. The incorporation of GGBS into jarosite waste also enhanced its durability. With higher GGBS percentage and longer curing time, the loss in both types of strengths caused by the alternative freeze and thaw cycles was reduced. Besides, the product stabilised with a higher concentration of GGBS and was more durable [58]. Gupta and Prasad [57] attempted to improve the geotechnical characteristics of jarosite utilising lime. Lime functions as an activator, thus improving its strength. Their investigation revealed that increased lime concentration and curing period increased the unconfined compressive and split tensile strength.

5.2 As a concrete material

Industrial waste material named jarosite still needs adequate research to be classified as a potential cementitious material. The literature shows that some jarosite-incorporated concrete has a positive effect on mechanical and durability properties. Mehra et al. [38], Gared and Gaur [39], Ray et al. [40], Saini et al. [41, 42], Gupta and Sachdeva [44], Debbarma et al. [46], studied the properties of concrete incorporated with jarosite as a partial replacement material for cement and fine aggregates, based on the observation made by the author's jarosite replacement percentage ranges from 5-25%, the mechanical and durability properties of the concrete tends to decrease beyond 25% replacement of jarosite. Though reports related to the use of jarosite as a concrete material are limited, the results on their improved mechanical and physical properties indicate a promising scope for utilising this zinc industrial by-product as a concrete material. However, further investigations need to be conducted to identify the long-term behaviour of jarosite when incorporated in concrete. Based on the observation from the previous studies, the incorporation of jarosite as a concrete material is a sustainable approach to reutilise the zinc industry waste and to reduce the consumption of cement in concrete also the mechanical and durability properties of the jarosite incorporated concrete have a slight increment in the performance which is explained in detail in the following sections.

6. Hydration process of jarosite

The hydration process of cement involves the formation of calcium hydrate gel as the outcome of the interaction between the orthosilicate ions (SiO^{4-}) of tri and dicalcium silicate. Upon further curing, the C-S-H gel matures and enhances the strength of the mix [89]. Ettringite, a needle-shaped crystal produced by the interaction of ferrite with tricalcium aluminate, and mono-sulfate are other components in the cement hydration process [90]. Equations 1-4 express the reactions that occur during the process of cement hydration.

Tricalcium aluminate + gypsum+ water → Ettringite +Heat

$$C_3A + 3CSH_2 + 26H \rightarrow C_6AS_3H_{32} \quad (1)$$

Tricalcium silicate + water → Calcium silicate hydrate +lime +Heat

$$2C_3S + 6H \rightarrow C_3S_2H_3 + 3CH \quad (2)$$

Tricalcium aluminate + ettringite + water → Monosulfate aluminate hydrate

$$2C_3A + 3C_6AS_3H_{32} + 22H \rightarrow 3C_4ASH_{18} \quad (3)$$

Dicalcium silicates + water → Calcium silicate hydrate + lime

$$C_2S + 4H \rightarrow C_3S_2H_3 + CH \quad (4)$$

From the reactions, it is evident that the hydration process releases heat. As per Ray et al. [40], due to jarosite's fine granularity when added to the cement mixture, it is expected to adhere to the cement grains, creating a barrier, as depicted in Figure 7. When the excess SO_3 in the jarosite interacts with water, SO_4^{2-} ions are released. These ions then react with the C_3A in the cement to produce ettringite, which forms a barrier on the surface of C_3S, thus reducing the water's ability to interact with cement and postponing the hydration process. Consequently, the more significant the jarosite in the mix, the longer the delay at the induction period. It was observed that C-S-H deficiency caused by mixing jarosite with cement could be primarily attributed to the decrease in readily available silicates, which was further balanced by a secondary hydration reaction. However, the deficiency was severe enough to be compensated by the secondary hydration at high concentrations of jarosite [40].

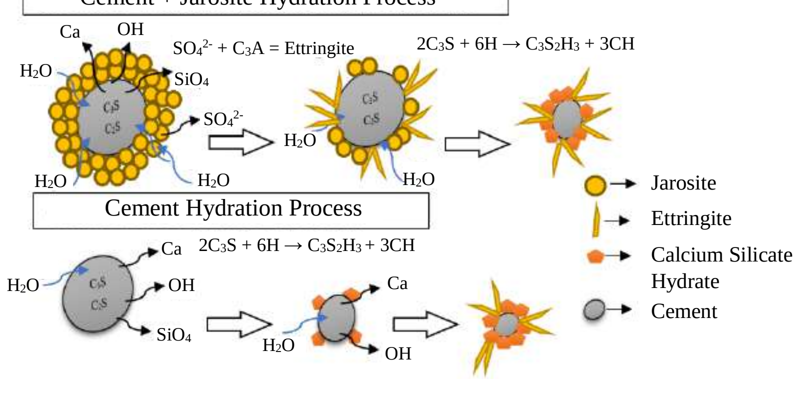


Figure 7 Schematic representation of the hydration process of cement and jarosite-blended cement.

7. Leaching characteristics of jarosite

The cumulative values of the Toxicity Characteristic Leaching Potential (TCLP) test on jarosite and jarosite incorporated construction material are presented in Table 3. The results indicate that raw jarosite obtained during the zinc leaching process can be extremely hazardous due to the presence of heavy metals, which pose a significant problem during the disposal of the waste material. However, when mixed with other materials, the proportion of heavy metal was drastically reduced, as illustrated in Figure 8. Among all the metal ions, copper was the least reduced metal ions from raw jarosite, with a reduction rate of 81%, while cadmium, with a reduction rate of 99.8%, was the most reduced. The textural and structural changes during sintering have significantly altered the jarosite product's mechanical, thermal, and chemical properties. The bulk of heavy metal oxides were discovered in low-soluble crystalline phases or glassy phases produced at higher temperatures, and this characteristic may help to decrease the possibility of harmful elements being leached out into the environment [2]. The hazardous elements were detoxified or immobilised during the sintering process under solid-state reaction through complexing in the silicate matrix [48]. The concentrations of nearly all the harmful components were discovered to be within the permissible limits, as advised by the United States Environmental Protection Agency (USEPA), for use in ecologically friendly applications. The sintered products from jarosite waste and clay had 15-45% CCRs [91]. Figure 8 represents the percentage reduction of toxicity of jarosite stabilised using various mixtures. Compared to the jarosite reference mix, the ppm values for the concrete mixes with added silica fume or fly ash were higher.

Table 3 Results of the toxicity characteristic leaching potential test of raw jarosite and jarosite-infused building materials

Sample			Heavy Elements(ppm)						Ref.
w/c	**% of jarosite**	**% of mineral admixture**	**Zinc**	**Lead**	**Copper**	**Cadmium**	**Iron**	**Silver**	
Raw jarosite		-	231.818	21.875	0.55	5.35	130	-	[38]
0.45	25	-	2.864	1.25	0.085	0.007	15	-	
Raw jarosite		-	-	35.87	-	27.33	-	78.54	[48]
0.27	75	-	-	0.694	-	0.314	-	0.248	
0.265	68	15(FA)	-	0.591	-	0.596	-	0.262	
Raw jarosite		-	-	35.87	-	27.33	-	78.54	[91]
0.220	42.5	15(CCR)	-	0.633	-	0.296	-	0.262	
Raw jarosite		-	237.55	26.03	0.493	4.982	110.85	-	[41]
0.38	15	-	20.486	1.15	0.098	0.105	18.32	-	
Raw jarosite		-	547	8.67	3.96	17.2	2.25	-	[44]
0.38	25	-	0.31	1.02	0.05	0.04	0.23	-	
0.5	20	-	8.25	0.2183	0.194	0.033	19.74	-	[40]
Raw jarosite		-	-	30.876	-	20.045	-	27.95	[58]
-	30	-	-	0.579	-	0.168	-	4.27	
-		-	231.81	21.875	0.55	5.35	130	-	[36]
0.45	25	Fine aggregate replacement	2.864	1.25	0.085	0.007	15	-	
Limits as per USEPA		**-**	**500**	**5**	**5**	**1**	**30**	**5**	**-**

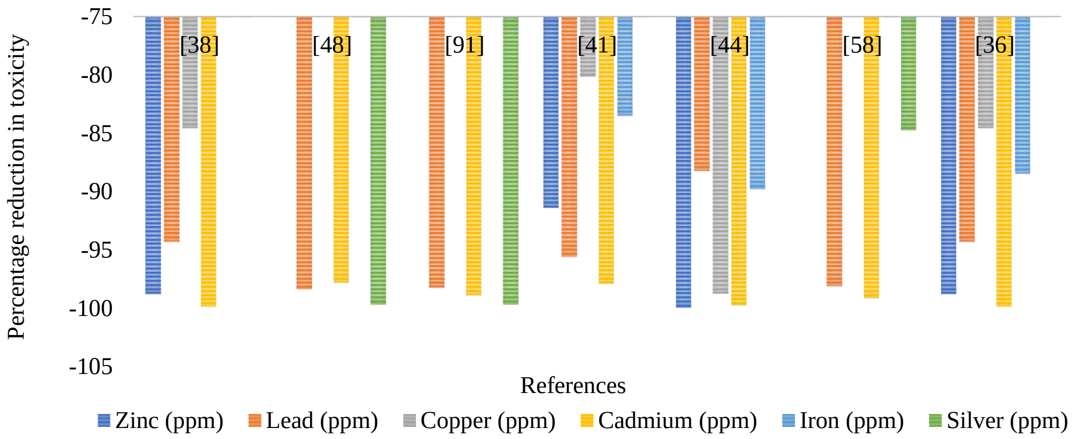


Figure 8 Percentage reduction of toxicity in heavy metals compared to raw jarosite when incorporated in concrete.

The variation in alkalinity could be the basis for this difference [41]. For all w/c ratios, the concrete mixes with the highest jarosite percentage (25%) appear to have reduced heavy metal leaching values [36]. Jarosite can be adopted as a building material as the heavy metal presence when incorporated in concrete is well below the USEPA limits.

8. Fresh concrete properties

For three different w/c ratios, Mehra et al. [92] investigated wherein the fine aggregate was substituted with jarosite content up to 25%. The authors reported that replacing fine aggregate with jarosite in the concrete did not affect the fresh concrete properties. The compaction factor ranged from 0.95- 0.98 for all the concrete mix ratios [92]. The maximum density of the concrete attained was 2874 kg/m^3 for 15% jarosite replacement for the highest w/c ratio of 0.5. For lower replacement levels of jarosite, the availability of C_3A content without sufficient SO_3 resulted in lower concentrations of ettringite formation, which could be the possible cause of the binding material's shorter setting time [42]. According to reports, up to 10% replacement levels did not affect the jarosite incorporated concrete mixes' setting times when compared to conventional mixes.

On the other hand, 20% replacement levels of jarosite resulted in delays of 70 and 50 minutes for the initial and final setting times, respectively [42]. The addition of jarosite in concrete does not affect the fresh concrete properties [43]. Thomas et al. [93] observed that by utilising copper slag, the workability of the concrete increased by 40%. Al-Jabri et al. [80] had a 205.66% increase in slump value for the total replacement of fine aggregate by copper slag. Wang et al. [94] reviewed the utilisation of copper slag in concrete and observed that the slump value increases with a higher replacement percentage of copper slag. [32] studied that there is a increase in 8-36% in workability when steel slag is replaced to 75% due to the effects of steel slag's fineness and high density. Compared to other industrial waste jarosite has the least effect on the workability of the concrete.

9. Compressive strength with jarosite as a binding material

The increase in compressive strength of jarosite incorporated concrete is caused by the highly amorphous nature and high specific surface area of the particles. With higher jarosite doses, the strength declines due to lower C_3A and C_3S contents due to decreased cement content [41]. The secondary hydration reaction of calcium hydroxide with silica generated a denser microstructure since fly ash, along with jarosite, comprises a significant quantity of silica, which is likely to cause a rise in strength over extended curing periods [41]. At higher fly ash dosage, the compressive strength drops since the secondary reaction cannot compensate for the deficiency of calcium silicate hydrate [41]. Gared and Gaur [39] reported that an increase in the jarosite proportion reduced the volume of voids, resulting in high density and improved strength parameters. Utilising Portland pozzolana cement (PPC)-43 grade cement in the mix resulted in better performance of jarosite incorporated concrete, producing increments of 7.38, 12.82, and 12.22 % in strength, whereas similar OPC utilised concrete [42] exhibited increases of 11.8, 8.68, and 10.52% respectively. This may be due to the presence of additional pozzolans and increased w/c ratio of the mix available for reaction during the secondary hydration by jarosite at later stages. This assertion is supported by previous research by [40] in which the concrete specimen exhibited good pore refinement and enhanced strength of 5.98 and 13.85% for OPC and PPC-based concretes, respectively. Although for lower w/c ratios, the compressive strength value decreased with the addition of jarosite, it was well within the desirable limits as interpreted by [43]. Figure 9 displays the percentage increment in compressive strength of the optimum mixes. The highest increments in the strength of 17.18 and 13.85 % have been observed for concrete containing PPC as the binding material and at high w/c ratios. Thus, it is evident that jarosite is more compactable with PPC cement blends than with OPC blends at high w/c ratios.

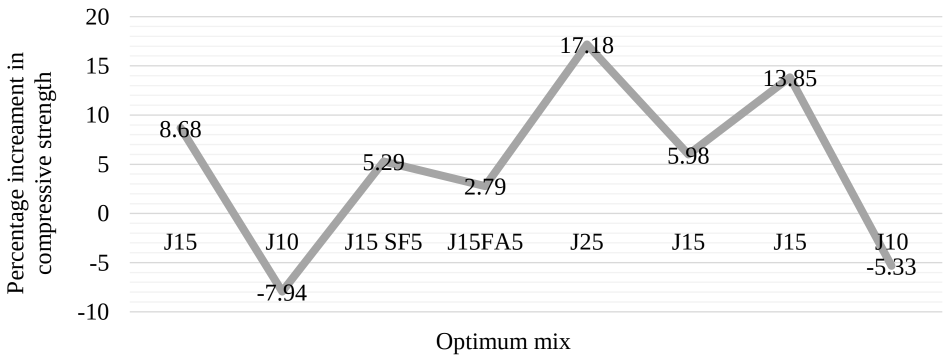


Figure 9 Percentage variation in compressive strengths of different concrete mixes [39-44]

A comparison of the compressive strengths of concrete incorporated with similar industrial wastes such as red mud [28, 95-97] and copper slag [98, 99] is presented in Figure 10. At a replacement level of 10%, RM showed an increase in compressive strength of 14.58%, whereas jarosite (JS) exhibited an increase of 11.03%. Copper slag also showed an increase in compressive strength with an increase in the replacement levels. However, with increasing replacement levels, the contribution of RM to the compressive strength followed a decreasing trend. On the other hand, JS exhibited a maximum increase in strength at 25%, as represented in Figure 10.

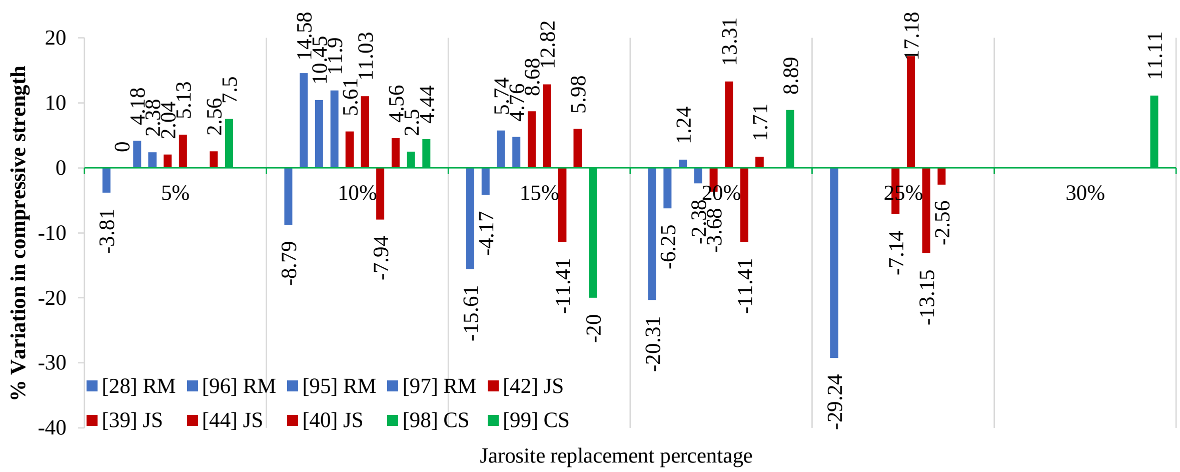


Figure 10 Comparison of compressive strengths of JS [39, 40, 42, 44], RM [28, 95-97] and CS [98, 99]

10. Compressive strength of concrete with jarosite as fine aggregate

Based on the research published on jarosite as a construction material, Mehra et al. [36] investigated the mechanical properties of conventional concrete by partially replacing the fine aggregate with jarosite. Among the literature databases collected, [36] and [38] are the only research studies to have utilised jarosite as a replacement for fine aggregate in concrete. Figure 11 displays the compressive strength behaviour of concrete in which the fine aggregate was partially replaced with jarosite up to 25% for three different w/c ratios of 0.4, 0.45, and 0.5. A noticeable increase in strength could be observed even at the early curing stages. The concrete mix with 25% replacement at 0.5 w/c ratio exhibited a 44% increase in compressive strength. This increase in compressive strength was compared with that produced by similar industrial waste materials such as copper slag, steel slag, lime slag [100], and quarry dust [100] used as an acceptable aggregate replacement, as shown in Figure 12. Jarosite behaves exceptionally well in terms of strength against its industrial waste counterparts. In the case of copper slag as fine aggregate, [93] observed that concrete density increased by 10.99% when fine aggregate was entirely replaced by copper slag. Comparatively, the compressive strength increased by 3.9% and decreased by 0.99% for 40% and 100% replacements with Copper slag, respectively. This interpretation corroborates with another study [101] wherein the total replacement of fine aggregate with CS resulted in an 11.17% decrease in strength. [102-104] explained the approach of using fillers as non-reactive waste material to improve the concrete properties. [100] studied the change in concrete parameters with different industrial waste fillers such as limestone dust and quarry dust that replace the conventional fine aggregate. It was observed that the optimum percentage for both quarry dust and limestone dust was 15 %, for which the corresponding increase in strength was 10.3% and 12.11%, respectively. [105] replaced the fine aggregate with steel slag up to 40% at a 10% increment rate. In this case, the optimum mix percentage was 20%, which produced a 35.04% increase in compressive strength, as illustrated in Figure 12. In addition to compression, replacing fine aggregate with steel slag improved the stiffness and brittleness of the concrete. Jarosite can be stated as a better fine aggregate replacement material based on the comparative analysis of the compressive strength parameter.

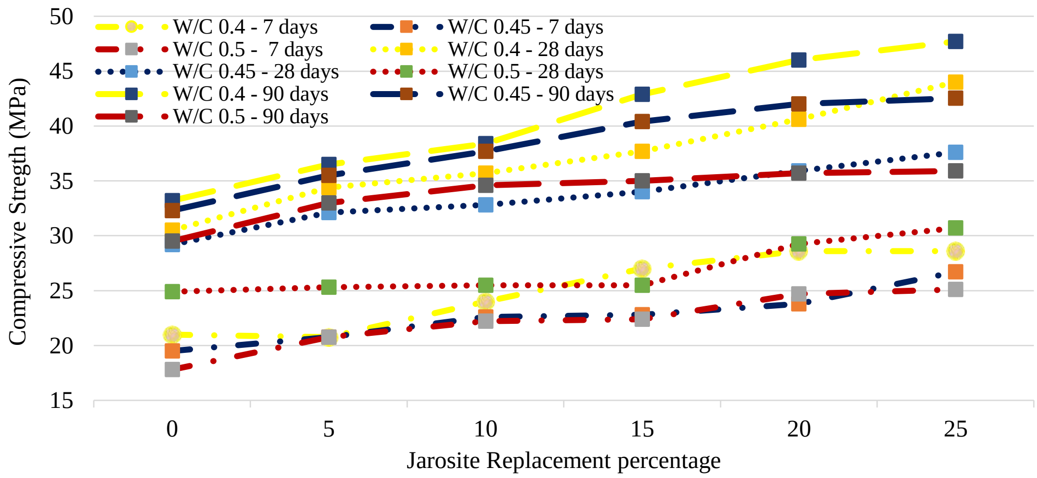


Figure 11 Variation of compressive strength of concrete with jarosite content as a fine aggregate replacement [36, 38]

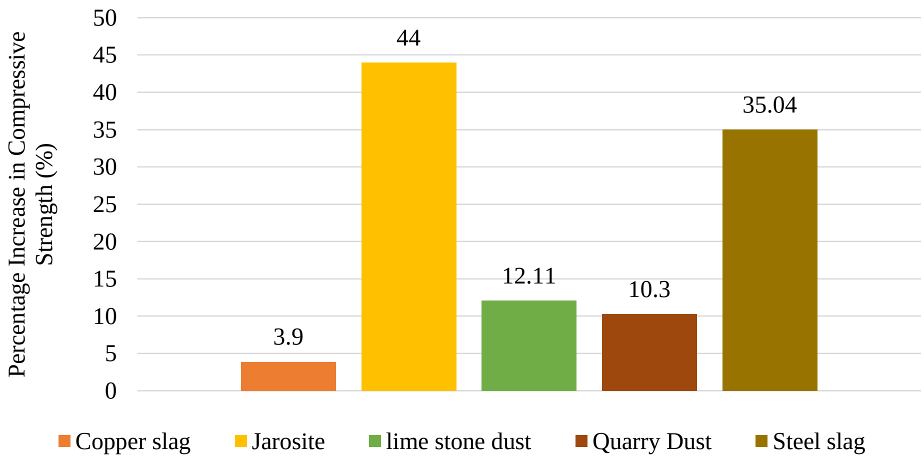


Figure 12 Percentage difference in the compressive strength of other fine aggregate replacement materials copper slag [80], jarosite [36], Lime stone dust [100], Quarry dust [100], steel slag [106]

11. Flexural strength of jarosite incorporated concrete

The flexural strength values correlate with the relative compressive strength values of the respective jarosite concrete mixes [36, 39, 41, 43, 44]. The inclusion of fly ash in jarosite concrete produced relatively lower values for flexural strength, while adding silica fume blends produced similar results with their compressive strength values. Nevertheless, both J15SF5 and J15FA5 concrete mixes exhibited higher values for flexural strength than the standard conventional mix [41]. After 28 days, J10, J15, J20, and J25 exhibited flexural strengths of 14.94%, 19.78%, 22.78%, and 28.4%, respectively, lower than the control mix [44]. Jarosite-incorporated concrete had a positive influence on compressive strength as the curing age increased, which is due to the effect of the secondary hydration process, as discussed in the previous section. In the case of flexural strength, this phenomenon is not applicable as the flexural strength tends to remain the same as the curing is increased to 180 and 365 days. The flexural strengths of the concrete using similar materials like red mud [28, 107, 108] and copper slag [98, 99] were compared to that of jarosite-incorporated concrete and presented in Figure 13. The literature has reported that concrete is weak at flexure with a significant negative variation in flexure strength. JS exhibited a maximum increased flexural strength of 15.32 % at a 20% replacement level. In contrast, RM had a maximum increase in strength of 24% at a 10% replacement level, and CS produced a maximum strength of 38.89% at a 30% replacement level.

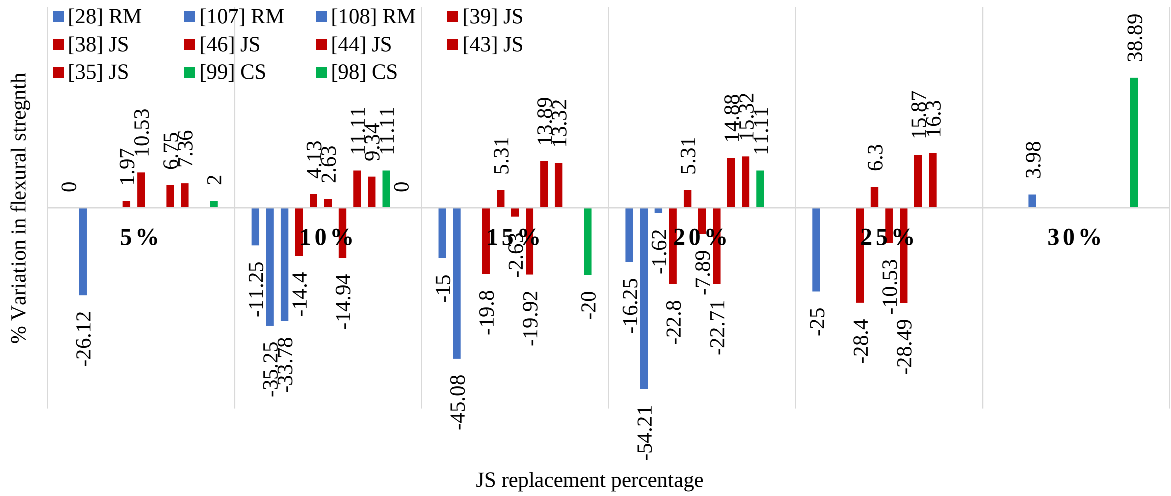


Figure 13 Percentage Variation in Flexural Strength of JS [35, 38, 39, 43, 44, 46], RM [28, 107, 108], and CS [98, 99]

Mathematical linear regression equations were formulated to correlate the flexural strength and jarosite replacement percentage from Figure 14. The variation in flexural strength obtained from the previous literature showed mixed trends. Flexural strength was shown to rise linearly with jarosite content, with a positive linear relationship formulated by linear regression models in [35, 38, 39].

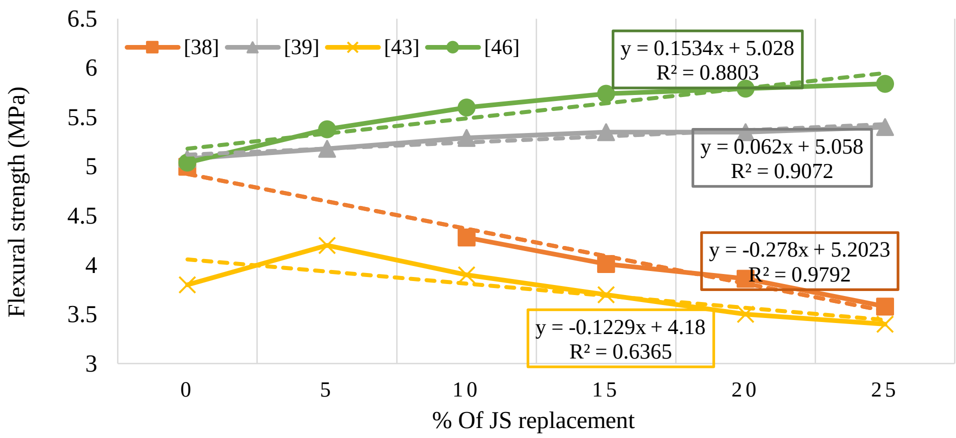


Figure 14 Correlation Between Flexural strength and JS Replacement percentage: data source [38, 39, 43, 46].

On the other hand, the linear regression models of [43, 44, 46] projected a linear decrease in flexural strength, which was in agreement with the variation of compressive strength of the corresponding concrete mixes. CS provided a positive increment in flexural strength compared to JS and RM. The R^2 values of all the mathematical models were more significant than zero and closer to 1. Thus, the jarosite replacement ratio proportionally affects the flexural strength of the concrete.

12. Water absorption of jarosite incorporated concrete

It has been established that the concrete's ability to absorb water declined as the jarosite proportion in the mix increased [39, 43], as is evident from Figure 15(a). Due to the fineness of the jarosite, the voids are reduced drastically, which reduces the permeability of the concrete, resulting in reduced water absorption. As a result, the J25 concrete mixes have the lowest water absorption percentage. Debbarma et al. [46] reported increased water absorption percentage when jarosite was incorporated with recycled asphalt pavement aggregate (RAP) concrete. The boost in water absorption is attributable to the hydrophilic character of the $FeSO_4$ phase in the jarosite particles, as well as water soaking the dust contaminants present in the RAP, which enhances the RAP's water absorption capacity [46]. Figure 15(b) reveals that the water absorption values for the J15SF15 and J15FA15 mixes are higher than those for the J15 mix but lower than that of the CM mix. Since silica fume and fly ash contain considerable quantities of silica, filling of the pores by the surplus C-S-H gel generated as an outcome of the secondary hydration reactions should have lowered the volume of permeable voids, which may have decreased the water absorption values of the concrete when compared to the CM and J15 mixes [41].

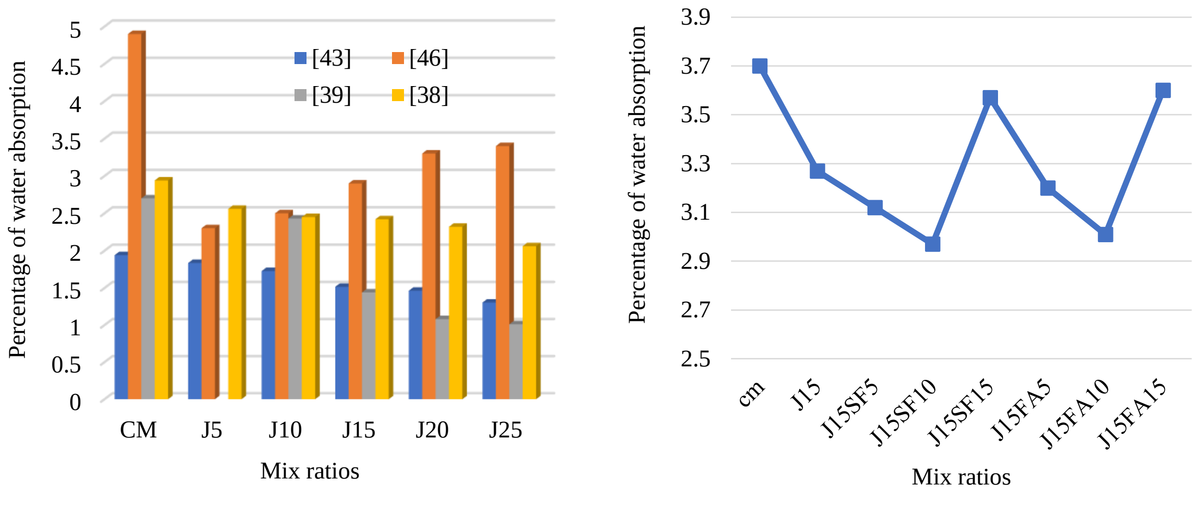
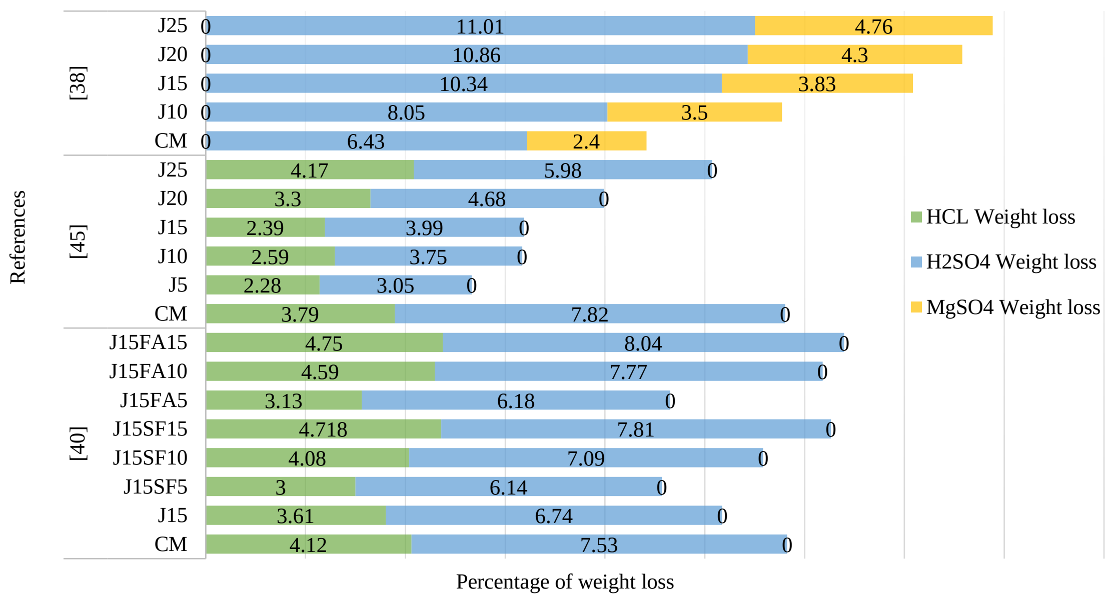


Figure 15 (a) Percentage of water absorption in jarosite-incorporated concrete [38, 39, 43, 46], (b) Percentage of water absorption of jarosite with SF and FA blends [41].

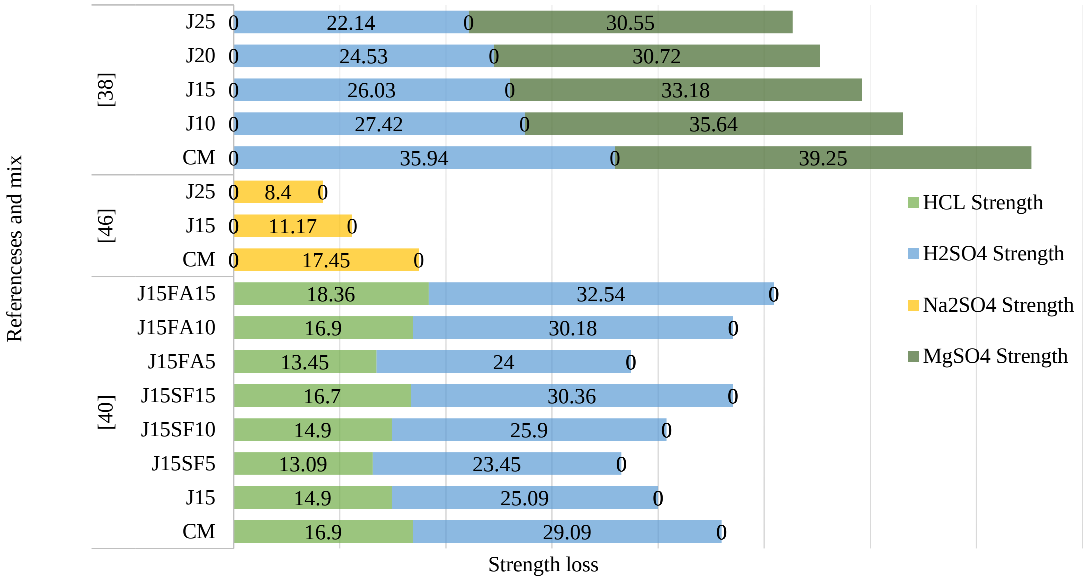
13. Acid resistance of jarosite incorporated concrete

The durability of the concrete is primarily based on its porosity. Interconnected pores in the concrete matrix generally attract the surrounding acidic ions, leading to concrete deterioration [46]. Owing to jarosite fineness, when incorporated into the concrete, it decreases its porosity, resulting in the formation of dense concrete. Saini et al. [41] observed that combining jarosite with additional binding material, such as silica fume and fly ash, provided different outcomes when immersed in hydrogen chloride (HCL) and sulfuric acid (H_2SO_4). The control and J15 control mixes were more vulnerable to acid attack than the J15SF5 and J15FA5 combinations. Mass loss and compressive strength reduction were slightly higher at more significant fly ash or silica fume replacement levels. In contrast to mixes developed with silica fume, a substantial decrease in strength was seen in fly ash mixtures [41]. Ray et al. [40] used Na_2SO_4 solution to cure concrete samples with 0, 15, and 25% jarosite replacement with OPC and PPC binders for 28 days. The result indicated weight loss in the concrete samples. The percentage reduction in compressive strength diminished as the jarosite content increased in both the OPC and PPC specimens [40]. Figure 16 represents the variation in the strength of jarosite incorporated concrete. This might be associated with aluminate depletion induced by jarosite's partial cement replacement. In contrast to jarosite, both OPC and PPC possess substantial aluminates. As more cement gets substituted by jarosite in the specimens, the number of aluminates available for reaction and, in turn, the generation of ettringite decline. The incorporation of jarosite also causes a secondary reaction involving silica and calcium hydroxide in the cement, limiting the quantity of calcium hydroxide required for gypsum precipitation. In general, adding jarosite to the concrete increased the resistance to acid and sulphate attacks by reducing the porosity of the concrete.



(a)

Figure 16 (a) Weight of jarosite incorporated concrete exposed to acid, (b) Strength loss of jarosite incorporated concrete exposed to acid.



(b)

Figure 16 (continued) (a) Weight of jarosite incorporated concrete exposed to acid, (b) Strength loss of jarosite incorporated concrete exposed to acid.

14. Abrasion resistance of jarosite incorporated concrete

The abrasion resistance of concrete is measured in terms of the depth of wear in accordance with IS 1237-1980. The recommended values for the maximum average wear for general-purpose tiles and heavy-duty tiles are 3.5 mm and 2 mm, respectively. Pavement abrasion resistance is a significant characteristic that provides a sense of the wear and tear that will be created on any pavement due to water seeping in and moving traffic. Abrasion resistance is equivalent to strength. After 28 days of curing, the mass loss for the J25 concrete mix was 0.5%, which was approximately 45% greater than for the standard mix and 30% and 22% higher than for the J10 and J15 mixes, respectively. The loss of mass caused by abrasion was reduced substantially for an extended period of curing. Compared to the mass loss after 28 days of curing, the mass loss of M40, J10, J15, J20, and J25 after 365 days was reduced by 37%, 43%, 49%, 45%, and 49%, respectively [44]. After seven days of curing, the J25 concrete mixture lost up to 0.554% of its weight, approximately 32% higher than the control mix. After 28 days of curing, the weight reduction had decreased to 0.387%, 0.443%, and 0.496% for J15, J20, and J25 mixes, respectively. The variations in the standard deviation were relatively small, illustrating that the abrasion loss was approximately the same for different combinations of mix ratios. As an outcome, increasing the jarosite content in concrete will not culminate in an exponential abrasion loss. [43].

15. Microstructural analysis of jarosite incorporated concrete

Jarosite powder possesses irregularly shaped, very fine particles, as discussed in section 4.1. This was also described by Ray et al. [40]. In the study conducted by [40], the morphological pattern of cement paste is observed along with 15 and 25% of jarosite incorporated cement paste. The densification of the microstructure is due to the secondary hydration caused by the presence of jarosite, and the fineness of jarosite grains also contributes to the densification by filling the microvoids in the concrete matrix. Gupta and Sachdeva [44] observed a high formation of ettringite formation with a high level of replacement percentage, also detected that for 10% and 15% replacement of jarosite incorporated concrete had low calcium to silica ratio than 20% and 25 % which indicates that there is a significant loss in polymerisation as the replacement level of jarosite is increased. Nandi et al. [109] observed that incorporating 15% of jarosite with OPC significantly improves the interfacial transition zone between aggregate and binder in the concrete matrix; the author also describes as the jarosite replacement level has increased, the concrete appears to be densely packed, the fineness of jarosite causes this phenomenon. Saini et al. [42] studied the morphology of cement paste with jarosite incorporation at 5-25%, by which the author detected that with a higher dosage of jarosite, a high volume of ettringite is observed, which results in low durability performance of the mortar. Saini et al. [41] studied the morphology of jarosite-incorporated concrete with fly ash and silica fume as mineral admixture based on the author's observation, denser particle packing was observed with 15% jarosite and 5% fly ash and silica fume; when compared to the morphology of control mix and concrete with only 15% jarosite, and also observed that the calcium by silica ratio is reduced when the curing days are increased, which indicates high polymerisation, which increases the strength of concrete. [110] stated that the high specific surface area may help to ascertain why C-S-H exhibits higher strength at a lower Ca/Si ratio. The aforementioned assertion was supported by observations in [41, 44, 110] that the compressive strength of the C-S-H paste improved when the Ca/Si ratio was reduced. Elevated concentrations of Si-O bonds were produced at a lower Ca/Si ratio [111]. The Figure 17 depicts the scanning electron microscope image of control concrete, 15% jarosite replaced concrete, and 25% jarosite replaced concrete. By the image, it is evident that the densification of the concrete matrix is observed as the jarosite replacement level is increased at 25 % jarosite replaced concrete, high ettringite formation is observed, and a high volume of calcium silicate hydrate gel is observed at concrete with 15% jarosite replacement. This supports the statements provided by various authors.

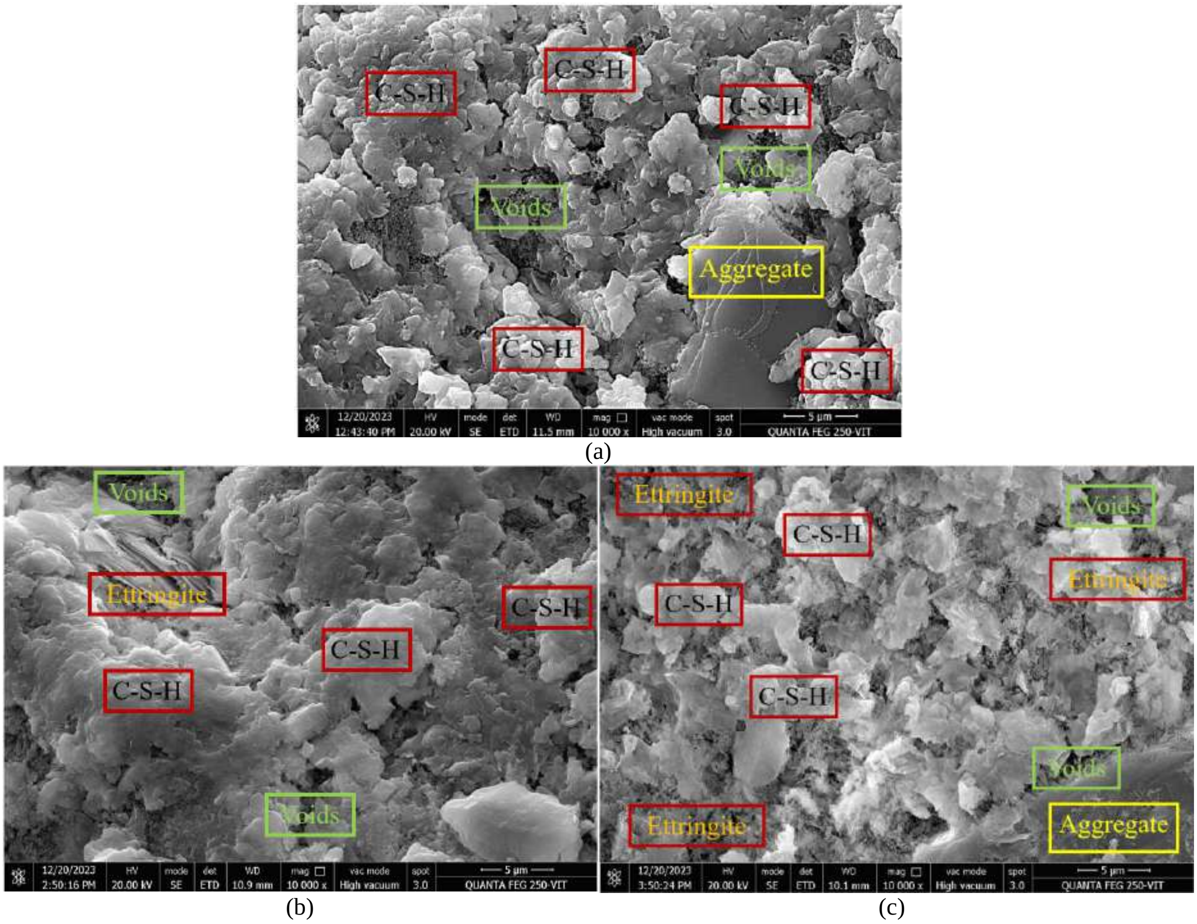


Figure 17 (a) SEM Image of Control mix, (b) SEM Image of 15% jarosite incorporated concrete, (c) SEM Image of 25% Jarosite Incorporated concrete.

16. Conclusion

The growth of the zinc industry in recent years has raised the need for sustainable disposal solutions for jarosite, a zinc industry by-product. Incorporating jarosite in the concrete solves various environmental issues, including reduced groundwater contamination and natural resource conservation. In this review, a comprehensive analysis was undertaken to determine jarosite-incorporated concrete's chemical, physical, mechanical, and durability characteristics. The findings of the review are listed below.

- As per ASTM standards, the proportion of the primary oxide constituents of jarosite ranges from 45.62 to 53.77%, due to which it can be classified as pozzolanic material.
- The particles of jarosite are finer than those in OPC, which prompts jarosite to fill the fine voids in the concrete, resulting in denser concrete.
- Incorporating jarosite in the concrete causes a delay of 70 and 50 minutes for the initial and final setting times, respectively. In contrast, the fresh concrete properties remain unaltered for jarosite blended cement concrete.
- The leaching potential of jarosite due to the heavy metal concentration had been a significant concern in utilising this material. Investigations reveal that blending raw jarosite with OPC, lime, and GGBS stabilises jarosite by drastically reducing its leaching potential by reducing the proportions of heavy metal ions. The TCLP values of the final product were within the limits of USEPA, thus providing a stable application for the hazardous material.
- The mechanical properties of the jarosite blended concrete indicate an increase in the strength characteristics of concrete incorporated with jarosite along with additional binders such as fly ash and silica fume.
- Jarosite can be incorporated into the concrete in two different alternative materials.
    - Jarosite as a binding material exhibited varying results but predominantly. A 15% replacement of jarosite as a cementitious material is determined to be the optimum mix proportion for maximum strength.
    - Jarosite as a fine aggregate replacement has enormous potential, mainly due to its increased strength at higher replacement levels. 25% replacement of jarosite as fine aggregate is the optimum mix proportion.
- The water absorption capability of the concrete decreased with an increase in jarosite content. The fineness of the jarosite reduces the microvoids in the concrete. The higher the jarosite content, the lower the porosity of the concrete, resulting in low water absorption. This property also provided enhanced acid resistance to the concrete. It has been found that the porosity decreases as the jarosite concentration increases in the concrete mixture, resulting in durable concrete.
- Abrasion resistance plays a crucial role in concrete application in pavements as it deals with the wear and tear of concrete surfaces. By incorporating jarosite, the abrasion resistance declined to a certain level, mainly due to the formation of excess ettringite caused by the secondary hydration process. Nevertheless, the reduced values of abrasion meet the requirement for pavement construction.

- The microstructural density of the concrete containing jarosite was enhanced, resulting in low pore volume and durable concrete.

This review indicates that jarosite possesses the chemical, physical, and mechanical properties to function as a viable concrete material. Various researchers suggest different optimum mix proportions, which may be attributed to adding mineral admixtures such as silica fume and fly ash. However, there needs to be more research available on the creep and shrinkage behaviour as well as the economic and environmental benefits of jarosite-incorporated concrete.

17. Recommendations

- Durability properties like dry shrinkage and creep parameters of jarosite-incorporated concrete need to be studied.
- Jarosite concrete as an electrically conductive concrete should be investigated thoroughly.
- A proper investigation into jarosite concrete's thermal conductivity and heat insulation properties is necessary.
- The durability parameters of jarosite concrete, such as abrasion and corrosion resistance, must be investigated.

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