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Environmental Impact Mitigation of Jarosite Waste Through Advanced Thermal Treatment Techniques

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Abstract: The latter approach is becoming more popular due to numerous advantages, such as possibilities for resource recovery from waste materials, cost-effectiveness, decreased landfill utilisation, and a reduced impact on the environment. Notably, waste compounds that contain metals like lead (Pb), copper (Cu), zinc (Zn) and cadmium (Cd) frequently exceed allowable limits and cannot be dumped in landfills or aquatic bodies. To ensure that these wastes satisfy legal requirements before their disposal or transform them into products with additional value, treatment of that waste is essential prior to disposal. Therefore, treating jarosite to reduce metal concentrations to acceptable levels or convert it into a value-added product is critical. To fulfil this need, jarosite waste is used directly in this study. The appropriate waste management improved thermal processing technique is adopted for copper, cadmium, zinc, and lead treatments to mitigate their impact on the environment. These approaches are becoming more popular because of their waste management benefits. This study focusses on incorporating jarosite waste in cement concrete, both directly and by thermal processing. Research shows that hydrometallurgical and pyrometallurgical wastes are valuable sources of metals. This study aims to analyse the leaching behaviour of jarosite in concrete before and after thermal treatment. Six experiments were carried out, using 15% jarosite waste (normal and heated at 600°C) in M30-grade concrete. The Toxicity Characteristic Leaching Procedure (TCLP) test determined the amounts of Zn, Pb, Cd, and Cu. The findings show that the metal concentrations ensure the suitability of jarosite for concrete production in both forms. This study assessed the chemical, physical, and microstructural features of concrete produced with jarosite as a partial cement substitute, with a focus on heat of hydration and durability. The results showed that untreated jarosite used in concrete produce best results, indicating that it can be used effectively to mitigate waste disposal challenges and reduce treatment costs.

Keywords: Jarosite, natural resources conservation, sustainability, thermal behaviour, toxicity characteristic leaching procedure (TCLP).

Introduction and Background

Various industrial processes produce liquid and solid waste, including aluminium, copper, and zinc compounds, posing substantial environmental risks due to the continuous generation of hazardous waste. The continuous production of hazardous waste, such as liquid effluents and solid wastes, raises environmental challenges. Notably, jarosite solid waste contains elements such as Cd, Zn, Pb, and Cu that exceed allowed limits, rendering direct disposal in landfills dangerous and non-compliant. Thermal processing, chemical treatment, and biological remediation are popular ways of dealing with industrial waste to reduce environmental impact and promote sustainable waste management practices. Researchers (Gared et al., 2024a; Mehra et al., 2018) have investigated how direct land filling of jarosite waste reduces the leaching of harmful

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Abbreviations

The following symbols/notations are used in this research paper.

AAS	Atomic Absorption Spectroscopy
Cd	Cadmium
CO_2	Carbon Dioxide
Cu	Copper
EPA	Environmental Protection Agency
HCl	Hydrochloric Acid
MTPA	Million Tonnes Per Annum
Pb	Lead
SDGs	Sustainable Development Goals
TCLP	Toxicity Characteristic Leaching Procedure
UNDP	United Nations Development Programme
ZHE	Zero Headspace Extractor
Zn	Zinc

pollutants like cadmium, zinc and lead. Although some researchers (Smith et al., 2006) have heated jarosite samples to 600°C before land filling to investigate leaching effects in soil, no studies have examined the impact of combining jarosite with concrete prior to land filling. Furthermore, the effects of heat treatment prior to and after absorption into concrete have not been studied. This study could significantly minimise the leaching of hazardous chemicals, hence reducing the likelihood of pollutant seepage into groundwater.The pyrometallurgical treatment used in jarosite thermal processing for zinc production optimises conditions for the least amount of lead and zinc in the slag and improves gas-liquid interactions for efficient metal removal (Rämä et al., 2018). Jarosite is a waste product of the hydrometallurgical zinc production process, and it is stabilised using the Jarofix technique prior to being deposited in landfills. Jarosite and alunite-type compounds, known for their stability and adaptability, are valuable in extractive metallurgy, environmental remediation, and planetary science, aiding in metal recovery and hazardous element control (Cruells and Roca, 2022). Utilisation of jarosite waste in concrete enhances sustainability in the construction industry by reducing CO_2 emissions, conserving natural resources, minimising energy, and lowering pollution by reducing the cement demand and diverting waste from landfills. It also helps to mitigate environmental pollution, reduces production costs, improves strength and durability, and makes it a valuable material for construction.

India is the 7th largest producer of zinc, with two zinc smelters located in Rajasthan. The Debari Zinc Smelter in Rajasthan produces zinc using the hydrometallurgical process, releasing hazardous by-product jarosite (Gared et al., 2024a). The Debari Zinc Smelter produces approximately 48 MTPA of zinc, leading to a significant amount of waste. This waste poses disposal problems in the smelter's landfills and has adverse effects on the environment, contaminating water, air, and soil (Gared et al., 2024b). Proper reuse and recycling of jarosite in the preparation of sustainable concrete can provide long-term benefits in terms of waste management and environmental impact reduction (Mehra et al., 2018). Jarosite is a basic hydrous sulphate of potassium and ferric iron (Fe-III) with a chemical formula of $KFe_3(SO_4)_2(OH)_6$ (Smith et al., 2006). It is a sulphate mineral that forms in ore deposits by the oxidation of iron sulphides. Jarosite is often produced as a byproduct during the purification and refining of zinc and is also commonly associated with acid mine drainage and acid sulphate soil environments (Smith et al., 2006). Jarosite usually appears in amber-yellow or dark brown colour and it has a trigonal crystal structure. Jarosite has a hardness grade ranging from 2.5 to 3.5 on the Mohs scale. It is mostly insoluble in water but rapidly dissolves in hydrochloric acid (HCl), demonstrating different solubility qualities depending on the media. Jarosite crystals are usually pseudo-cubic or tabular but can also be found in granular crusts, nodules, fibrous masses, or concretionary forms. It is brittle and has a distinct basal cleavage (Smith et al., 2006). Jarosite waste powder is represented in Figure 1.

Figure 1: Jarosite waste in powder form.

Jarosite as Industrial Waste

Jarosite waste is produced by zinc electrolytic processing industries and has a direct negative impact on the environment (Ali et al., 2014; Mehra et al., 2016a). The disposal of jarosite sludge is a major environmental issue, as it is categorised as hazardous waste due to its high concentrations of toxic elements such as iron, lead, zinc, sulphur, cadmium, chromium and copper (Mehra et al., 2016b). In addition to the toxic elements, jarosite waste often contains other essential metals.There has been an increased focus on reprocessing jarosite waste to recover valuable products in recent years (Gared and Gaur, 2021). It can be converted into useful materials such as nano-particles, pigments, filling materials, acidic reagents, adsorption materials, catalytic materials and construction materials. Mombelli et al. (2021) assessed the mechanical and metallurgical properties of briquettes formed from a combination of jarosite powder and blast furnace waste (Mombelli et al., 2021). When a binder was added to the briquetting process, the mechanical resistance qualities of the briquettes improved. However, non-uniform heating of the powder resulted in the creation of an undesired chemical, which had a negative impact on the mechanical qualities of the briquette (Mombelli et al., 2021). Demir et al. (2008) explored the metal leaching behaviour of hazardous zinc industry waste before and after thermal treatment (Demir et al., 2008). The samples were examined using the TCLP and ASTM techniques.The addition of clinoptilolite, blast furnace slag, and red mud to zinc leach residue dramatically reduces the heavy metal concentration of the effluent. In addition to red mud outperforming both clinoptilolite and blast furnace slag at 1200°C, Zn^{2+} levels were obtained at 0.025 mmol/L (Demir et al., 2008). XRD, SEM, and TG/DTA investigations were performed to assess the properties of jarosite waste, identify potential environmental implications, and comprehend its recycling or usage potential. These techniques were employed to characterise the jarosite waste properly. TCLP testing indicated that harmful materials leaching from jarosite waste contained 5625.21 mg/kg of zinc, 131.54 mg/kg of lead, and 58.67 mg/kg of cadmium. Islam et al. (2021) investigated the usage of jarosite (JS) as a filler material in pavement construction. The performance of asphalt mixtures containing JS was investigated for various forms of distress and their environmental appropriateness. The research demonstrated that asphalt mixes containing JS were highly resistant to cracking, moisture sensitivity, and rutting (Islam et al., 2021). The jarosite's high porosity, smaller particle size, large specific surface area, and strong asphalt-filler interaction all contribute to its exceptional performance. A study presented the properties of jarosite and fly ash and investigated their potential usage as construction materials. The incorporation of particular ratios of jarosite and fly ash into bricks and concrete revealed that these materials could achieve compressive strengths that meet industrial standards. The findings demonstrate that, in the right quantities, jarosite and fly ash can be employed successfully in construction applications.

Bibliometric Analysis

Figure 2A illustrates a line graph of the number of jarosite-related publications in Scopus digital database from 2000 to 2024. This trend confirms a general increase in research focus on jarosite and its waste utilisation to reduce disposal problems over time. Therefore, Figure 2(B) shows a co-occurrence network map developed by VOSviewer computer software that

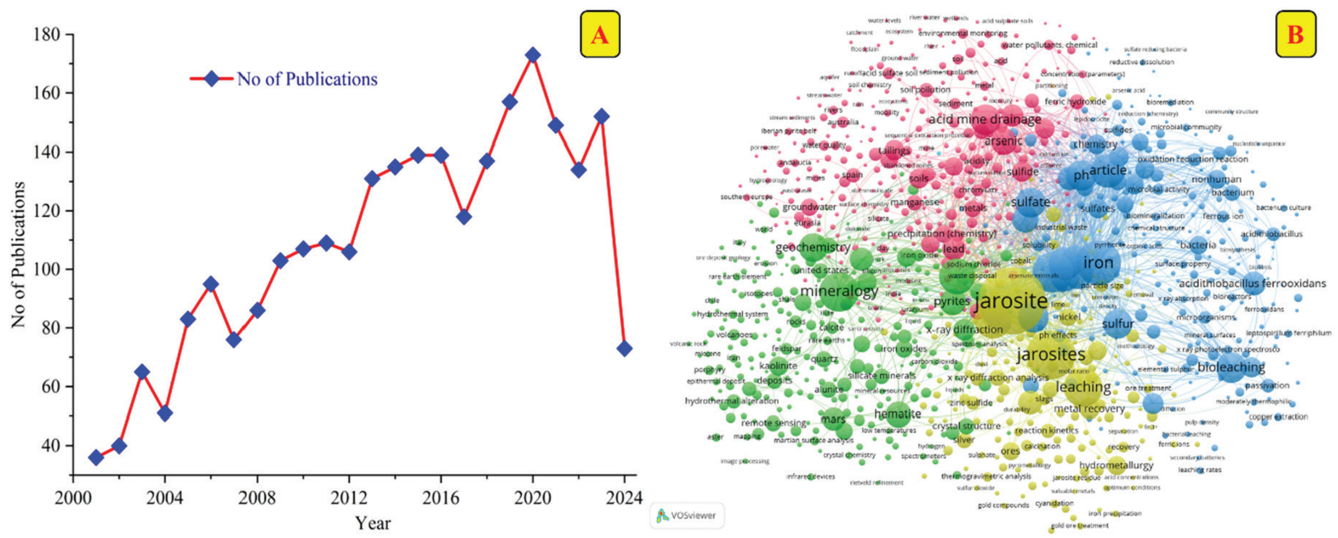

Figure 2: Scopus database based (A) research trend and (B) co-occurrence diagram.

depicts the relationship and frequency of keywords associated with jarosite research (Khichad et al., 2024). The map is colour-coded, with clusters reflecting various topic areas of the research field. The keyword "jarosite" occurs centrally, showing its importance in the investigations.

These graphics demonstrate the expanding interest and research focus on jarosite, as well as the various areas of study that are related to it.The existing available literature shows a lack of studies on the properties of Jarosite-based concrete before and after thermal treatment. There is also a need for wide-ranging assessments of jarosite's chemical, physical, and durability properties, as well as a need to study the leaching behaviour of jarosite to recommend precise outcomes.

Objective and Importance of the Study

The main objective of this study is to investigate the leaching behaviour of jarosite-containing concrete. This study also analysed the chemical, physical and microstructure characteristics of the jarosite waste. Additionally, the jarosite feasibility after and before heat treatment were assessed by heating at 600°C temperature at various stages. This study provides a significant solution to the use of jarosite waste in concrete production and reduces the demand for cement. It reduces the disposal issue of jarosite waste and saves the cost involved in treatment through the Jarofix technique as per government regulations for hazardousness reduction prior to the disposal stage. Moreover, reduced demand for cement reduces CO_2 emissions, minimises energy, conserves natural resources, reduces concrete production costs, and improves strength and durability in the production of eco-friendly, sustainable, non-hazardous concrete.

Materials and Methodology

This section describes the materials and experimental methodologies used to explore the integration of jarosite waste in concrete and assess the impact of this practice on the mechanical characteristics and overall performance of the concrete mixes, including the effect of thermal treatment at 600°C. The methodology followed in the investigation of leaching behaviour is shown in Figure 3.

Sample Preparation

This study aims to find the concentration of Zinc (Zn) and Lead (Pb) present in the jarosite powder and in the cubes made with the normal and heated jarosite powder. Here, for completion of the aim, Jarosite is taken as 15% of the weight of cement for cube construction. Here, six different cases are made using jarosite powder as heated and normal along with cube formation for further studies shown in Table 1.

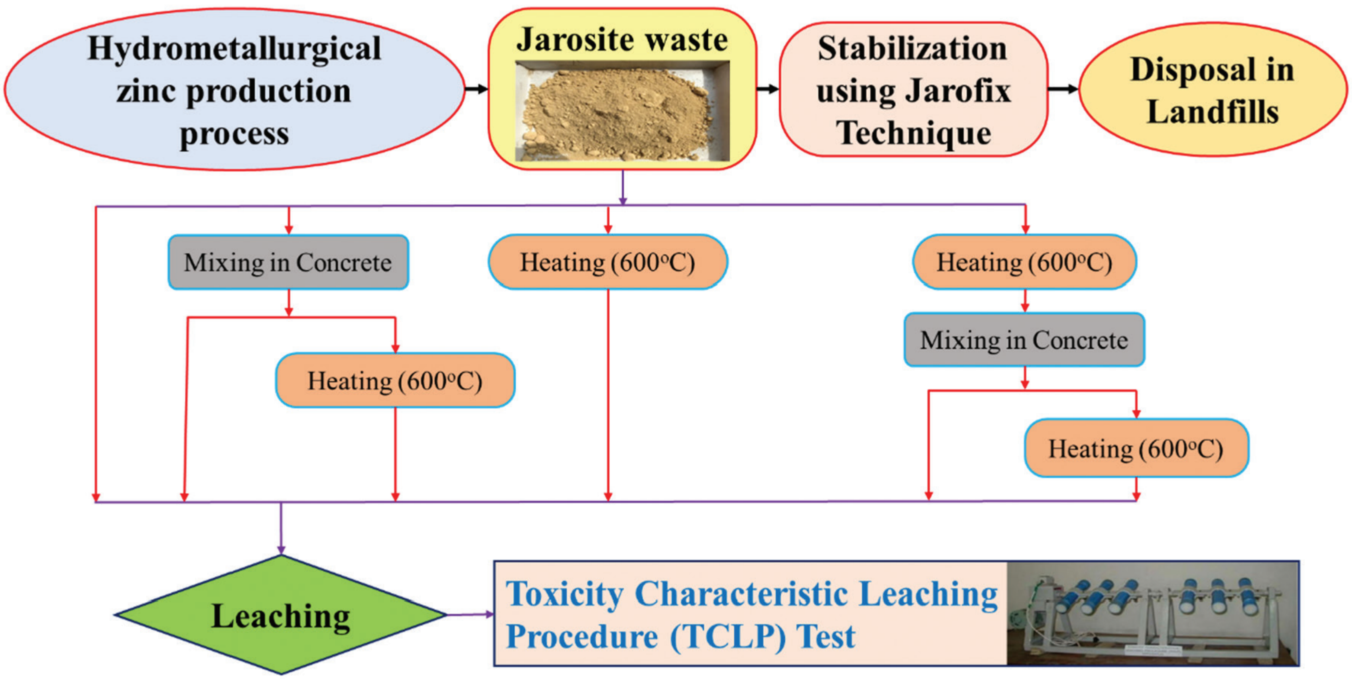

Figure 3: The methodology followed to perform TCLP tests on Jarosite.

Table 1: Six different cases for analysis of jarosite leaching behaviour

Case No	*Jarosite heating*	*Treatment before TCLP*
I	No	Normal Jarosite Powder
II		Normal Jarosite in Concrete
III	Yes	Heated at $600^{O}C$ Jarosite Powder
IV	(600ºC)	Heated at $600^{O}C$ Jarosite in Concrete
V		Normal Jarosite in Concrete + Heating at $600^{O}C$
VI		Heated Jarosite in Concrete + Heating at $600^{O}C$

Testing of Raw Materials

The basic characteristics of the raw materials used in this study were ascertained and their suitability for usage in concrete mixtures was confirmed by testing them for bulk density, specific gravity, and consistency adhering to the standard guidelines as shown in Figure 4.

The bulk density of river sand (Zone-II) and fine aggregates (10 mm and 20 mm) in loose and dense state is shown in Table 2.

Table 2: Density calculation of the aggregates in different states

Aggregate	*State*	*Density (gm/cc)*
Fine	Loose	1.418
	Compacted	1.586
10 mm	Compacted	1.479
20 mm	Loose	1.575
	Compacted	1.700

Preparation of Concrete

In this study, M30 grade of concrete cubes were casted with jarosite substituting 15% for the cement content to investigate leaching behaviour.

A maximum nominal aggregate size of 20 mm was employed in the concrete of M30 grade with quantities of raw materials shown in Table 3. Sand (from zone-II), 10 mm and 20 mm aggregates all recorded water absorption rates of 0.57%, 0.42%, and 0.88%, respectively. A slump value of between 25 and 50 mm was obtained by setting the initial water-cement ratio at 0.5. The net water content was 199.997 grams after accounting for the aggregates' water absorption, yielding a corrected water-to-cement ratio of 0.54.

Leachate Preparation

For the purpose of maintaining a 1:10 ratio, 20 grams of the sample was initially added to 200 ml of deionised water. Subsequently for an entire day, the beaker was mounted up on a rotatrimeter. The material was subsequently permitted to remain undisturbed for a further 24 hours. Afterwards this, Watman filter paper no. 42 was used to vacuum filter the sample. The sample was then filtered and run through an Atomic Absorption Spectroscopy (AAS) machine for the detection of the heavy elements, as shown in Figure 5.

Toxicity Characteristic Leaching Procedure (TCLP)

The Toxicity Characteristic Leaching Procedure (TCLP) examines the ease with which inorganic and organic contaminants can migrate through different types of

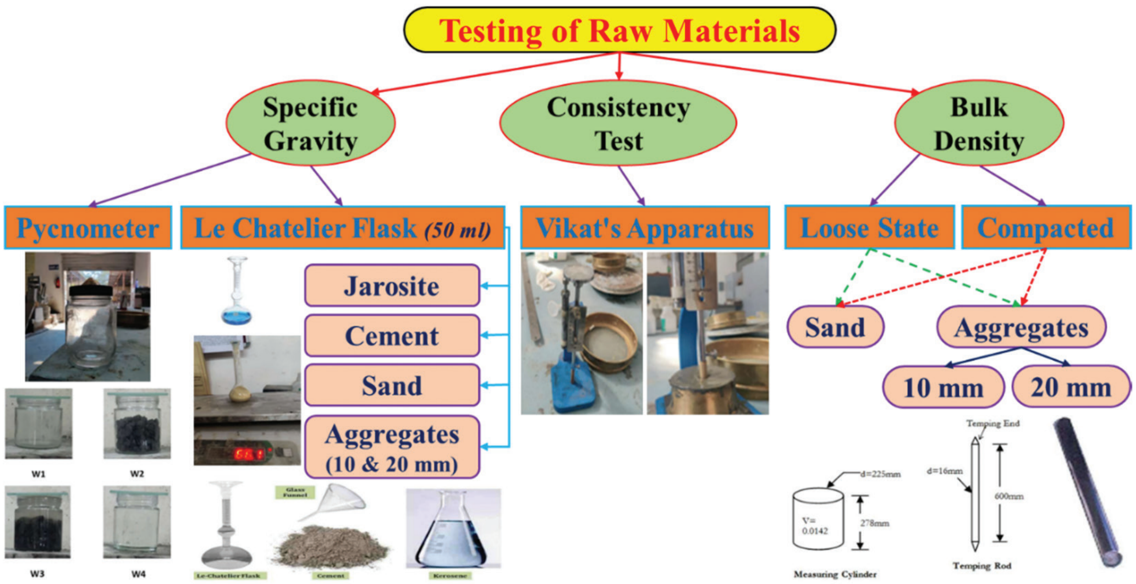

Figure 4: Various tests performed on the raw materials.

Table 3: Mix design input raw materials adhering to IS:10262

Materials	*Specific Gravity*	*Mass (gm)*	*Volume for 1 m^3 Concrete (m^3)*	*Corrected Mass (gm)*
Cement	3.18	372	0.11698	372
Sand	2.608	680.8643	0.38	677.005
Water	1	186	0.186	199.997
10 mm aggregates	2.747	585.0456	0.62	579.942
20 mm aggregates	2.71	577.1655		572.131
Admixture	NA	0	0	0

waste, such as liquids, solids, and multiphasic materials (Figure 6). There is no requirement for the TCLP test if a study reveals that certain compounds are either absent or present at levels below US Environmental Protection Agency regulations. The waste is considered hazardous and additional examination of other fractions is not needed if the TCLP liquid fraction includes any substances listed by the USEPA at or above regulatory limits. The waste is dangerous and the Zero Headspace Extractor (ZHE) is not needed if the bottle extractor analysis shows that any controlled volatile analysis exceed regulatory levels. Furthermore, it is not possible to guarantee that the volatile component levels in the mineralized water container's liquid are within legal limits.

The Toxicity Characteristic Leaching Procedure (TCLP) aims to measure the concentration of heavy metals content in a specific filter media or sludge before its under coagulation in the disposal stage. Moreover, it also determines the aqueous heavy metal content in these materials using the EPA-developed TCLP method.

Figure 5: Heavy metals detection on atomic absorption spectroscopy machine.

Figure 6: TCLP agitation apparatus.

Results and Discussion

Results of Specific Gravity Tests

Material tests are conducted on different elements to determine their corresponding values and to use them in the cube-casting process. Figure 7 shows the specific gravity of tests performed on the materials, including aggregates, cement and jarosite.

Results of Normal and Heated Jarosite Samples

The zinc (Zn) concentration decreases from Case-I to Case-II and subsequently increases from Case-II to Case-III, as shown in Table 4. Additionally, it is

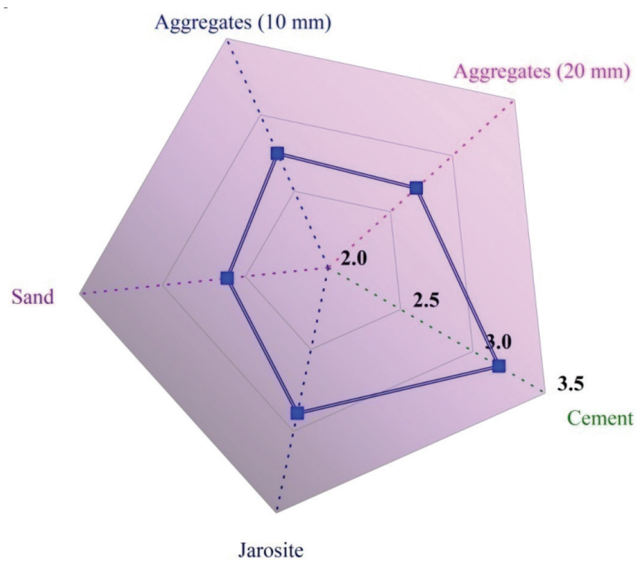

Figure 7: Specific gravity of raw materials.

evident that the copper (Cu) concentration falls between Cases-I and II before rising between Cases III and IV. The findings show that the cadmium (Cd) concentration falls between Cases I and II, then rises between Cases II and III. The lead (Pb) concentration rises from Case-I to Case-II and then again from Case-II to Case-III, according to the data. Hence, it can be seen that Case-II (Normal Jarosite in Concrete) is superior to Case-III (Heated Jarositein Concrete) and Case-I. The results also shows that the concentration of zinc reduces from Case-IV to Case-V, while subsequently increases from Case-V to Case-VI. The concentration of copper also observed decrease from Case-IV to Case-V, and subsequently increases from Case-V to Case-VI. The results indicate that the concentration of cadmium falls from Case-IV to Case-V, and subsequently increases from Case-V to Case-VI. Therefore, the concentration of lead increases from Case-IV to Case-V, then from Case-V to Case-VI. So, it reveals that Case-II (Normal Jarosite in Concrete) is a better alternative.

From the concentration trends throughout the cases shown in Figure 8, it is observed that Case II has the lowest concentration, followed by Cases-V, III, VI, I, and IV. Focussing on the concentration of components in jarosite-incorporated concrete, the order is Case-II, Case-V, Case-III, and Case-VI. When regular jarosite powder is compared to heated jarosite powder, the former gives larger concentrations (Case-I > Case-IV). Moreover, the investigation shows that zinc, copper and cadmium concentrations are reasonably high in powder form samples, but lead concentrations are significantly greater in jarosite-incorporated concrete.

Table 4: Concentration of metals in different cases of jarosite

Case	*Description*	*Metals Concentration in ppm*			
		Zn	*Pb*	*Cd*	*Cu*
I	Normal Jarosite Powder	2.782	0.307	0.192	1.866
II	Normal Jarosite in Concrete	0.041	0.52	0.005	0.084
III	Heated Jarosite in Concrete	0.067	0.641	0.049	0.711
IV	Heated Jarosite Powder	3.71	0.4	0.13	1.338
V	Normal Jarosite in Concrete + Heating	0.046	0.55	0.007	0.099
VI	Heated Jarosite in Concrete + Heating	0.123	1.31	0.012	0.162

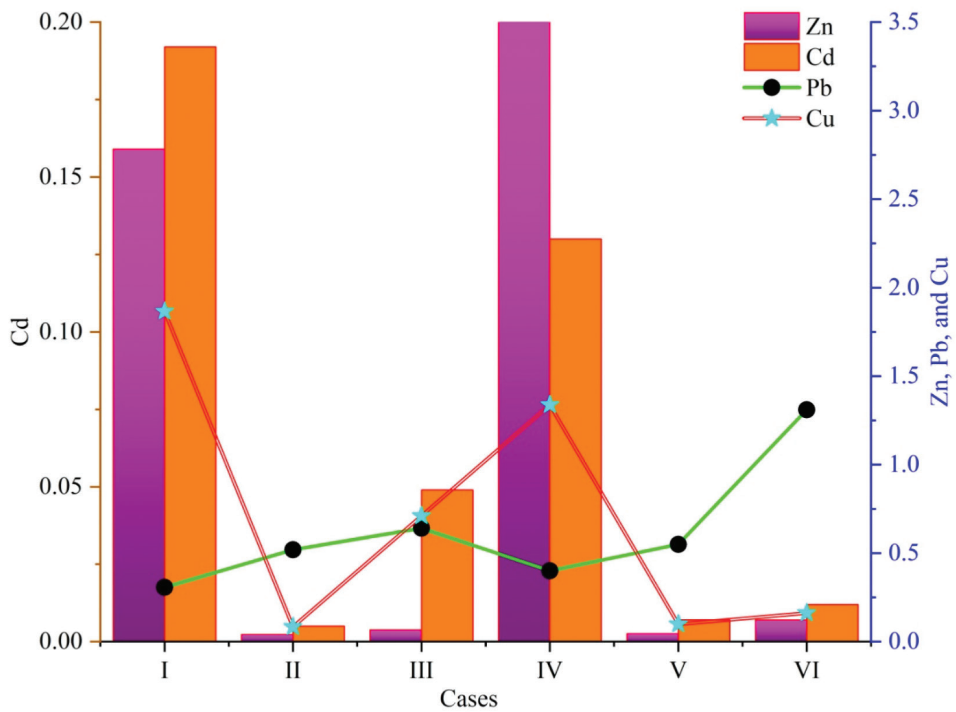

Figure 8: Metallic concentrations (ppm) in various jarosite samples.

Sustainable Development Goals

This experimental research aligns the incorporation of jarosite waste in concrete to five key Sustainable Development Goals (SDGs) as per UNDP (United Nation Development Programme) Agenda 2030. It promotes industrial innovation and sustainable infrastructure (SDG 9), reduces CO_2 emissions and improves urban environmental quality (SDG 11), reuses industrial waste and reduces environmental impact (SDG 12), mitigates climate change by reducing CO_2 emissions (SDG 13), and prevents hazardous waste pollution while maintaining ecosystems (SDG 15). Comparably, numerous studies found in literature broadly in areas of novel materials, pavements, maintenance and construction, etc. support the SDGs. Overall, the study reveals an entire approach to sustainable development that prioritises resource efficiency and pollution reduction as shown in Figure 9 (United Nation Development Programme, 2015).

Conclusions and Recommendations

This study investigated the leaching behaviour of jarosite in concrete, comparing untreated and thermally treated samples across multiple cases. Generally, untreated samples showed superior performance compared to thermally treated ones. Incorporating jarosite into concrete enhanced its leaching behaviour. Among the cases studied, mixing normal jarosite in the concrete (Case-II) yielded the most favourable results. This recommends that using this method for concrete casting is advisable. The overall trend based on heavy metal concentrations across all cases is as follows: Case-II > Case-V > Case-III > Case-VI > Case-I > Case-IV. Specifically, for cases involving concrete made with either normal or heated jarosite, the trend is: Case-II > Case-V > Case-III > Case-VI. Comparing normal jarosite powder and heated jarosite powder, the trend shows: Case-I > Case-IV.

The chemical characterisation indicated higher zinc concentrations in powder form, whereas lead concentrations were notably higher when mixed into concrete. This pilot study highlights the importance of further comprehensive investigations, including detailed chemical, physical, and microstructural characterisations, to provide more robust recommendations for practical applications in the future. For improved results and decision-making, expanding chemical characterisation to include various other heavy metals is recommended. Additionally, analysing the physical, chemical, and microstructural properties of jarosite provides essential insights into its behaviour. For more accurate determination of leachate elements in TCLP tests, using more concrete cubes crushed into powder would enhance precision in future studies.

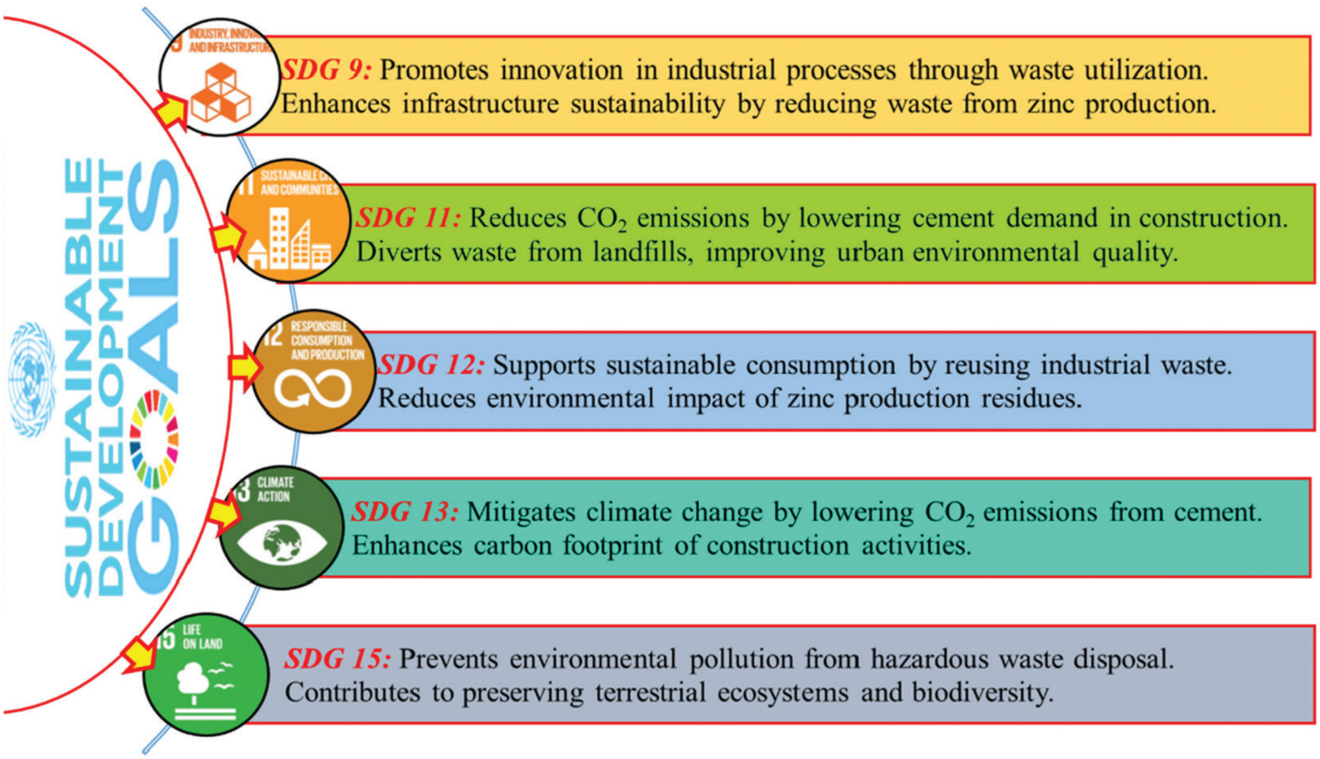

Figure 9: Aligned SDGs of the study as per UNDP.

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Ethical approval
This article did not contain any experiments with animal or human subjects carried out by any of the authors.

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