CHARACTERIZATION OF WATER TREATMENT SLUDGE AND ITS IMPLICATIONS ON CONCRETE AS A PARTIAL REPLACEMENT OF CEMENT

By

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A thesis submitted in partial fulfilment of the requirements for the degree of MASTER of SCIENCE in CIVIL ENGINEERING.

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Declaration

I hereby declare that the work contained in this Thesis has not been previously submitted to meet requirements for an award at this or any other higher education institution. To the best of my knowledge and belief, the Thesis contains no material previously published or written by another person except where due reference is cited. Furthermore, the Thesis complies with PLAGIARISM and ACADEMIC INTEGRITY regulation of CUET.

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Dedication

To my respected teachers

and

To my beloved parents

List of Publications

[As per the decision from the 139th Academic Council on 06/02/2022, Decision 139/1 (amended from 136/7 decision 2): "From the academic year 2020-2021, the student has to publish one (01) international publication (peer-reviewed) journal/conference for awarding MS/ M.Sc./ M. Engg./ M. Phil. degree. The student is required to publish two papers in Scopus/ISI/Thomson Reuters-indexed journals to obtain the Ph.D. degree]

Journal Article

- **K. Arsh**, B. C. Mondal, M. H. Masum, "Characterization of water treatment sludge and its implications on concrete and cement mortar as a partial replacement of cement", *Construction and Building Materials*, 458 (139454), 1-21, https://doi.org/10.1016/ j.conbuildmat.2024.139454.
- **K. Arsh**, B. C. Mondal, M. F. R. Zuthi, "Evaluation of Life Cycle Assessment (LCA) and Heavy Metal Leaching from Concrete with Partial Replacement of Cement by Water Treatment Plant Sludge (WTS)"(Draft prepared).

Conference Paper

- **K. Arsh**, B. C. Mondal, "Exploring the Potential of Water Treatment Sludge (WTS) as Cementitious Materials and Its Application in Cement Mortar as a Partial Replacement of Cement" *2nd International Conference on Mechanical, Manufacturing and Process Engineering (ICMMPE – 2024)*, Dhaka, 29-30 May, 2024, 396-402.

Declaration by the Supervisors

This is to certify that **Kandila Arsh** has carried out this research work under our supervision, and that she has fulfilled the relevant Academic Ordinance of the Chittagong University of Engineering and Technology, so that he is qualified to submit the following Thesis in the application for the degree of MASTER of SCIENCE in CIVIL ENGINEERING. Furthermore, the Thesis complies with the PLAGIARISM and ACADEMIC INTEGRITY regulation of CUET.

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Abstract

Application of supplementary cementitious materials (SCMs) in concrete is advantageous as it reduces the need for cement production, thereby lessening the environmental carbon footprint. However, this practice is restricted the release of contaminants from concrete into the environment. This study aims to investigate the physical, chemical, morphological, and mineralogical properties of WTS and evaluate its feasibility as a partial cement replacement in cement mortar and concrete. Moreover, this study evaluates the environmental impacts of cement production with optimal clinker replacement by WTS through heavy metal leaching and Life Cycle Analysis (LCA). Cement mortar and concrete specimens were prepared with various WTS addition levels, and tested for different properties of concrete and mortar. The leaching behavior of heavy metals was also investigated, while the LCA was performed using the cradle-to-grave method. The study found that WTS particles are irregular with an average size of 9.20 μm. WTS has high SiO_2, Al_2O_3, and Fe_2O_3 content which meet the pozzolanic standards. The cement mortar with the mix ratio of 1:2 and WTS of 7.5% shows the highest compressive strength compared to other mix ratios. In addition, flexural strength peaks at 5% WTS but continues to drop beyond this limit. However, compressive strength of the concrete is increased up to 5% WTS addition while, the unit weight decreases with higher WTS, ranging from 2160-2567 kg/m^3. The leaching concentration trend of partially replaced concrete at 63 days was as follows: Cu > Mn > Ni > Cr > Fe > Cd. The differences in leaching concentration between the control and partially replaced concrete were determined to be 1.2-2.8 times higher for the 1:2:4 ratio and 1.3-3.6 times higher for the 1:1.5:3 ratio. However, concentrations stabilize after 40 days and stay within regulatory limits. The study also found that replacing 5% clinker with WTS in cement production reduces the carbon footprint by 1.91% and shows a reduction in various other environmental impact categories by 0.09% to 3.31%. This study demonstrates that WTS can effectively be used as a partial cement replacement, offering environmental benefits and improved mechanical performances at optimal levels.

বিমূর্ত

কংক্রিটে পরিপূরক সিমেন্টিয়াস পদার্থের প্রয়োগ বিভিন্ন দিক বিবেচনায় সহকারী ভূমিকা পালন করে, কারণ এটি সিমেন্ট উৎপাদনের প্রয়োজনীয়তা কমায়, ফলে পরিবেশগত কার্বন ফুটপ্রিন্ট হ্রাস পায়। তবে, এই পদ্ধতির সীমাবদ্ধতা হল কংক্রিট থেকে পরিবেশে দূষণকারী পদার্থের নির্গমন, যা পরিবেশ এবং মানবস্বাস্থ্যের জন্য সম্ভাব্য বিভিন্ন সমস্যার কারণ হতে পারে। এই গবেষণার উদ্দেশ্য হলো WTS-এর ভৌত, রাসায়নিক, আকৃতিগত, এবং খনিজগত বৈশিষ্ট্যগুলি বিশ্লেষণ করা এবং কংক্রিট ও সিমেন্ট মর্টারে আংশিক সিমেন্ট প্রতিস্থাপন হিসাবে এর ব্যবহারযোগ্যতা মূল্যায়ন করা। এছাড়াও, এই গবেষণায় ভারী ধাতুর নির্গমন এবং জীবনচক্র বিশ্লেষণ (LCA) এর মাধ্যমে WTS দ্বারা ক্লিংকারের সর্বোত্তম প্রতিস্থাপনের ক্ষেত্রে সিমেন্ট উৎপাদনের পরিবেশগত প্রভাব মূল্যায়ন করা হয়েছে। বিভিন্ন পরিমাণে WTS সিমেন্টের পরিবর্তে যুক্ত করে কংক্রিট এবং সিমেন্ট মর্টারের নমুনা প্রস্তুত করা হয়, যার মাধ্যমে নমুনাসমূহের সক্ষমতা, শক্তি, এবং ওজনসহ কংক্রিট ও মর্টারের আকৃতিগত এবং খনিজগত বৈশিষ্ট্যগুলো পরীক্ষা করা হয়েছে। গবেষণায় দেখা গেছে যে WTS কণাগুলি অনিয়মিত এবং এদের গড় আকার ৯.২০ মাইক্রোমিটার। WTS-এ উচ্চ পরিমাণে SiO_2, Al_2O_3, এবং Fe_2O_3 রয়েছে, যা পোজ্জোলানিক মানদণ্ড পূরণ করে। ৭.৫% WTS সহ ১:২ মিশ্রণ অনুপাতের সিমেন্ট মর্টার অন্যান্য অনুপাত এর তুলনায় সর্বোচ্চ সংক্ষেপণ শক্তি প্রদর্শন করে। নমনীয় শক্তি ৫% WTS-এ সর্বোচ্চ হয়, কিন্তু এই সীমা অতিক্রম করার পর ক্রমাগত কমতে থাকে। WTS-এর ৫% সংযোজন পর্যন্ত কংক্রিটের শক্তি বৃদ্ধি পায় এবং এর পরিমাণ বৃদ্ধির সাথে সাথে একক ওজন কমে যায়, যা মিশ্রণ অনুপাতে ২১৬০- ২৫৬৭ কেজি/মি^{3} এর মধ্যে থাকে। ৬৩ দিনের পর আংশিক প্রতিস্থাপিত কংক্রিটের নির্গমন ঘনত্বের প্রবণতা ছিল নিম্নরূপ: তামা > ম্যাঙ্গানিজ > নিকেল > ক্রোমিয়াম > আয়রন > ক্যাডমিয়াম । কন্ট্রোল এবং আংশিক প্রতিস্থাপিত কংক্রিটের মধ্যে নির্গমন ঘনত্বের পার্থক্য ১:২:৪ অনুপাতে ১.২-২.৮ গুণ এবং ১:১.৫:৩ অনুপাতে ১.৩-৩.৬ গুণ বেশি পাওয়া গেছে। তবে, ৪০ দিন পর ঘনত্ব স্থিতিশীল হয় এবং নিয়ন্ত্রক সীমার মধ্যে থাকে। এছাড়া, গবেষণায় দেখা গেছে যে সিমেন্ট উৎপাদনে ক্লিংকারের ৫% WTS দ্বারা প্রতিস্থাপন করলে কার্বন ফুটপ্রিন্ট ১.৯১% হ্রাস পায় এবং অন্যান্য বিভিন্ন পরিবেশগত প্রভাবের ক্ষেত্রে ০.০৯% থেকে ৩.৩১% পর্যন্ত হ্রাস প্রদর্শিত হয়। গবেষণার ফলাফলগুলি দীর্ঘমেয়াদী পরিবেশগত প্রভাব নির্দেশ করে এবং সর্বোত্তম কর্মক্ষমতা এবং ন্যূনতম পরিবেশগত প্রভাব সহ কংক্রিট তৈরিতে সহায়তা করে।

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Nomenclature

AAS	: Atomic Absorption Spectrometry
ACI	: American Concrete Institute
AR	: Alumina Ratio
ASTM	: American Society for Testing and Materials
BDS	: Bangladesh Standards
BECR	: Bangladesh Environmental Conservation Rules
BSTI	: Bangladesh Standards and Testing Institution
CA	: Coarse Aggregate
C-S-H	: Calcium Silicate Hydrates
CV	: Coefficient of Variation
DIN-EN	: Deutsches Institut für Normung - Europäische Norm (German Institute for Standardization - European Standard)
EDX	: Energy-Dispersive X-ray Spectroscopy
FA	: Fine Aggregate
GGBS	: Ground Granulated Blast Furnace Slag
GHG	: Greenhouse Gas
LCA	: Life Cycle Assessment
LCI	: Life Cycle Inventory
LCIA	: Life Cycle Impact Assessment
LSR	: Lime Saturation Ratio
MK-SS	: Mann–Kendall and Sen's slope
OPC	: Ordinary Portland Cement
PCA	: Principal Component Analysis
PCs	: Principal Components
PVC	: Polyvinyl Chloride
SCM	: Self-Compacting Mortar
SCMs	: Supplementary Cementitious Materials
SEM	: Scanning Electron Microscopy
SPSS	: Statistical Package for the Social Sciences
SR	: Silica Ratio
UPV	: Ultrasonic Pulse Velocity
USEPA	: United States Environmental Protection Agency
USGS	: United States Geological Survey
WTS	: Water Treatment Sludge
XRD	: X-Ray Diffraction
XRF	: X-Ray Fluorescence

Chapter 1: INTRODUCTION

1.1 GENERAL

The introduction chapter highlights the environmental challenges posed by cement production and sludge management. Cement manufacturing significantly contributes to global CO_2 emissions, and the disposal of water treatment sludge (WTS) in developing countries results in pollution and environmental hazards. The study aims to address these issues by exploring WTS as a partial replacement for cement in concrete and mortar, assessing its potential benefits and environmental impact. The scope includes testing WTS in various mix ratios and evaluating its effects on material properties, while also assessing heavy metal leaching and conducting a life cycle assessment. Limitations include a focus on specific mix ratios and curing periods, with no long-term performance data or analysis of other materials and environmental impacts.

1.2 ENVIRONMENTAL IMPACTS OF CEMENT PRODUCTION

The cement industries are emitting substantial CO_2 which significantly exacerbates climate change, contributes to air pollution, and impacts overall environmental health due to its large carbon footprint. Cement is responsible for 7% of global greenhouse gas (GHG) emissions, with concrete emitting 347 to 351 kg CO_2 equivalent per cubic meter at a 0.5 w/c ratio [1,2]. It is reported that the cement industry emits an average of 222 kg eq. CO_2 for the production of one ton of cement [3]. In addition, The cement production caused a sharp increase in global CO_2 emissions from 0.157 to 1605 million metric tons within the duration of 1960 to 2022 [4,5]. Global cement production hit 4.1 billion tons in 2023, resulting in approximately 910 million tons of CO_2 emissions, with the industry emitting an average of 222 kg CO_2 Eq. per ton of cement [3,6]. If this trend continues, cement-related CO_2 emissions in developing countries, excluding China, are estimated to rise 1.4–3.8 gigatons by 2050 [7]. Researchers worldwide are seeking alternatives to cement to reduce its substantial environmental impact, focusing on reducing CO_2 emissions and promoting sustainable construction practices. Given these substantial emissions, the use of alternative cementitious materials in concrete is essential to reduce the environmental impacts of concrete production, lower greenhouse gas emissions, and promote more sustainable construction practices.

1.3 CHALLENGES AND ENVIRONMENTAL IMPACTS OF SLUDGE MANAGEMENT

The rapid population growth has led to an alarming increase in waste generation, posing global challenges for recycling industries, particularly in developing countries, as they grapple with contamination, climate change, and the adoption of green policies [8]. Every day, different types of sludge, including tannery, sewage, textile, water treatment plant, paper industry, electroplating, and domestic sludge, are generated. These sludges are typically discharged into the environment by being released into nearby water bodies, buried in nearby land, or taken to nearby landfill sites [9–12]. Discharging sludge into the environment can lead to water and soil pollution, harm aquatic and terrestrial ecosystems, and pose health risks due to contaminants [13,14]. The adverse effects and environmental challenges of sludges have led to the search for alternative uses. Water treatment plants in Bangladesh generate substantial amounts of sludge, which is typically discharged into nearby water bodies or dumped in open sites. Water treatment sludge, consisting of solid sediment and suspended solids (SS), is a by-product of water treatment plants. It is generated during various purification processes such as sedimentation, coagulation, flocculation, filtration, and disinfection. A single water treatment plant can produce WTS as much as 100,000 ton/year [15–17]. The generation rate of dry water treatment sludge per capita ranges from 0.98 to 6.42 ton/year [18]. However, a treatment plant in Chattogram generates approximately 40 tons of waste per month, totalling around 480 tons annually [19]. This sludge is often disposed of by releasing it into nearby water bodies or by dumping it in open landfill sites in many developing countries. Hence, managing the WTS has become a pressing global concern. The cost for WTS disposal ranges from $130 to $200 per ton, with total average costs reported at approximately $37.5 million in the Netherlands and $5.88 million in the UK [18,20,21]. The uncontrolled disposal of WTS contaminates soil and clogs subsurface pores, limiting water infiltration [10,22,23]. The major coagulants used in these plants are aluminum and iron salts ($Al_2(SO_4)_3.18H_2O$, $FeCl_3.6H_2O$, $FeCl_2$, and $FeSO_4.7H_2O$) [22,24]. Although this method (discharging and dumping of dry WTS) is cost-effective, there is a risk of contaminating water bodies and soil at dumping sites due to the high salt content if the WTS is not treated before disposal [22]. High concentrations of these salts can be toxic to aquatic life, and excessive alum salts can cause soil fixation, hindering plant growth [24,25]. Furthermore, WTS may contain heavy metals from the raw water sources and chemicals used in the treatment process [24,25].

1.4 PROBLEM STATEMENT

The adverse impacts and environmental challenges of WTS have prompted the search for alternative uses due to space constraints and hazards like groundwater contamination, air pollution, and land pollution. While various studies have shown that the quantity and properties of WTS vary with water quality, seasonal changes, and coagulant dosage [22,25,26], there is a significant gap in research specific to Bangladesh. Existing literature often focuses on isolated properties of WTS, but no comprehensive study has been conducted in this geographical context. This study will fill a critical gap in existing research by comprehensively characterizing water treatment sludge (WTS), focusing on its physical, chemical, morphological, and mineralogical properties. Despite some studies on WTS application in concrete as discussed earlier, there is limited research on its partial replacement of cement in cement mortar, particularly in the context of Bangladesh. The generation of WTS has become a significant environmental concern in Bangladesh due to the increasing number of water treatment plants. Uncontrolled disposal of WTS into nearby water bodies or open sites leads to soil contamination and reduced water infiltration [10,22,23]. Moreover, WTS may contain heavy metals derived from the raw water and the chemicals employed in the treatment process [24,25]. The potential risks associated with the release of contaminants from concrete into the environment, particularly when exposed to water, can have serious implications for ecological systems. Moreover, limited studies [8,27–29] have investigated the potential environmental risks associated with various SCMs in concrete, and no studies have been found to date on the leaching of heavy metals from concrete with partial replacement of cement by WTS.

1.5 AIMS AND OBJECTIVES

The increasing focus on sustainability and environmental impact in the construction industry has led to the exploration of alternative materials that can reduce reliance on traditional resources. The specific objectives are as follows:

1) To investigate the physical, chemical, and morphological characteristics of Water Treatment Sludge (WTS).

2) To evaluate the efficiencies of WTS as an alternative for cementitious material.

3) To assess the environmental benefits of using WTS compared to conventional cement.

1.6 SCOPES AND LIMITATIONS OF THE STUDY

This study investigates the incorporation of water treatment sludge (WTS) into cement mortar and concrete, assessing its impact on material properties and environmental effects. Concrete specimens are prepared using two mix ratios: 1:2:4 and 1:1.5:3, with WTS replacing 2.5% to 15% of the cement. The concrete is cured for various periods—7, 14, 28, and 72 days—to evaluate how different curing times affect compressive strength. For cement mortar, mix ratios of 1:2, 1:4, and 1:6 is tested, with both compressive and flexural strengths being measured. Leaching tests are performed solely on concrete specimens to analyze the concentrations of heavy metals—Chromium (Cr), Copper (Cu), Iron (Fe), Cadmium (Cd), Manganese (Mn), and Nickel (Ni)—over a period of up to 63 days [30]. The study also includes a life cycle analysis (LCA) to compare the environmental impacts of traditional Ordinary Portland Cement (OPC) and OPC with a 5% clinker replacement by WTS, considering 18 different environmental impact categories.

The study has several limitations that affect the breadth and depth of its findings. It focuses on only two specific concrete mix ratios, potentially limiting the applicability of the results to other mix designs used in practice. Compressive strength is the only performance metric evaluated for concrete, with no assessment of flexural or tensile strengths, which are also important for a comprehensive evaluation. The curing periods are fixed and may not represent all possible scenarios, and long-term performance beyond the curing periods is not explored. For cement mortar, only three mix ratios are tested, and no long-term performance data is provided. The leaching tests are confined to concrete, excluding other materials like mortar, and only a specific set of heavy metals is analyzed. The environmental impact assessment is limited to a 5% clinker replacement and considers 18 impact categories, which may not encompass all potential environmental effects or lifecycle stages of cement production.

1.7 THESIS OUTLINE

Chapter 1: Introduction outlines the environmental challenges associated with cement production and water treatment sludge (WTS) disposal, discussing the study's aim to explore WTS as a partial cement replacement and assess its benefits and environmental impact.

Chapter 2: Literature Review provides a review of using waste materials in concrete and mortar, discussing their benefits, challenges, and the chemical and physical properties of WTS, including leaching characteristics.

Chapter 3: Materials and Methodology describes the materials and methods used, including characterization of raw materials, specimen preparation, statistical analyses, leaching tests, and life cycle impact assessment.

Chapter 4: Results and Discussion presents the findings on binder characterization, the impact of WTS on concrete properties, heavy metal leaching, and life cycle environmental impacts.

Chapter 5: Conclusion summarizes the potential of WTS as a sustainable cement alternative, noting improvements in workability and strength at lower replacement levels while addressing challenges such as strength reduction and heavy metal leaching at higher levels. The chapter concludes with recommendations for careful integration of WTS and further research on long-term durability and environmental safety.

Chapter 2: LITERATURE REVIEW

2.1 GENERAL

This chapter provides a comprehensive review of using waste sludge materials as partial replacements for cement in concrete and mortar. It discusses the potential benefits and challenges of incorporating these materials, including their availability, environmental impact, and effects on concrete properties. The chapter also investigates how WTS functions as a cementitious material, exploring its chemical and physical properties that make it suitable for use in construction. Additionally, it evaluates the environmental impact of using supplementary cementitious materials by investigating the leaching characteristics of concrete containing WTS, assessing the release of heavy metals and other contaminants to determine their compliance with environmental standards.

2.2 WASTE SLUDGE MATERIALS AS PARTIAL REPLACEMENTS OF CEMENT

Researchers have been working from the last few decades to reduce the cement production by introducing some mineral admixtures having cementitious properties such as fly ash [31–35], glass powders [36–38], rice husk ash [39,40], wood ash [41,42], GGBS [43–45], etc. Researchers are still investigating to explore new alternative cementitious materials to reduce CO_2 emissions, construction waste generation, high energy consumption, natural resources depletion, habitat destruction, and air and water pollution [5,46–48]. Similar to mineral admixtures, the researchers investigated on various types of sludges such as sewage sludge [49–52], textile sludge [9,29,53,54], hypo sludge [55–57], paper waste sludge [58], and tannery sludge [12,59–62] etc.

Table 2.1 provides a comprehensive overview of the studies investigating the use of various waste sludge materials as partial replacements for cement in concrete. The replacement levels vary widely ranging from 0% to 50% with recommended proportions spanning from 5% to 25%. The compressive strengths for these recommended replacement levels range from 20.23 MPa to 61.24 MPa. Moreover, the table shows that the tensile strength of 2.85 MPa to 3.75 MPa and the flexural strength of 2.92 MPs to 5.91 MPa for the recommended replacement levels. The above mechanical properties of the composite concrete indicate that the studied sludges have potential cementitious properties

The proportion of cement content is higher in cement mortar compared to cement concrete. To reduce the cement content in mortar preparation, several studies were conducted on cement mortar made with partial replacement of mineral admixtures as shown in **Table 2.2**. Brick waste was reported to use as a partial substituted material in mortar preparation with a recommended proportion of 5-10%, resulting in a compressive strength of around 45 MPa [63,64]. Schackow et al. (2015) [65] and Singh & Garg (2006) [66] found that the fired clay brick and Indian clay can also be used as a partial replacement material with the maximum replacement level of 40% but the compressive strength was found significantly lower than other partially substituted cement mortars. Glass powders [67,68], granite powders [69] and ceramic powders [70] individually have been reported as potential substituted cementitious materials used in cement mortar with a recommended proportion varying 5-35% having maximum compressive and flexural strength ranging between, 21-49 MPa and 4.5-6.9 MPa, respectively.

Table 2.1. Studies focusing on various waste sludge materials as partial replacements of cement in concrete.

Partial re-placement materials	Proportion of replacement	Recommended proportion	Compressive Strength for recommended proportion (MPa)	Tensile/ Flexural strength for recommended proportion (MPa)	References
Sewage sludge with GGBS	7.5-15%	10.5%	39.91	3.75/3.54	[71]
Lime sludge with wollastonite powder	0-15%	10%	58.20	3.56	[60]
Waste paper sludge ash	0-30%	15%	51.50	-	[58]
Textile Sludge	0-50%	20%	39.97	-	[9]
Sewage Sludge Ash	0-20%	15%	22.70	2.72	[72]
Tannery sludge	0-25%	25%	36.81	-	[73]
Waste Paper sludge with Paper Sludge Activated Charcoal Powder	5-20%	10%	52.30	5.91	[74]
Waste Paper Pulp	2.5-15%	10%	36.87	-	[75]
Textile Sludge	0-20%	5%	44.50	4.02	[53]
Hypo sludge	0-40%	10%	61.24	3.10/5.78	[56]
Textile Sludge	0-30%	10%	20.23	2.85/3.32	[54]
Hypo sludge	0-10%	5%	43.30	2.97	[57]

Table 2.2. Studies focusing on various waste materials as partial replacements in Cement Mortar.

Partial re-placement materials	Proportion of replacement	Recommended proportion	Compressive Strength for recommended proportion (MPa)	Tensile/ Flexural strength for recommended proportion (MPa)	References
Fly ash	10-50%	30% and 50%	19.73 and 19.25 MPa	-	[76]
Blend of Marble and Granite Waste Powder	5-30%	10%	32.5 MPa	-	[77]
Brick powder	5-15%	5%	45.52 MPa	-	[64]
Waste Granite Powder	5-15%	5% to 10%	20 and 21 MPa	4.0-4.5 MPa	[69]
Recycled green glass powder	10-60%	Up to 35%	28.6-48.6 MPa	6.90-6.92 MPa	[67]
Ceramic Waste Powder	10-30%	Up to 20%	35-38 MPa	-	[70]
Textile sludge	5-25%	5%	44MPa	3.8 MPa	[53]
Ceramic Powder and Silica Fume	5% of Silica Fume and 10-20% of Ceramic powder	5% of Silica Fume and 15% of Ceramic	62.19 MPa	-	[78]
Glass Powder	5- 25%	5-10%	33.2-34.0 MPa	4.7 MPa	[68]
Fired clay brick waste	10- 40%	Up to 40%	5.5 MPa	-	[65]
Microwave Incinerated Rice Husk Ash (MIRHA)	5-25% for 1:3, 1:4, 1:5, 1:6 ratios	Up to 20%	14-36 MPa for 1:3 ratio, 5- 18 MPa for 1:4 ratio, 3-20 MPa for 1:5 ratio, 3-20 MPa for 1:6 ratio	-	[79]
Sewage sludge ash	10-30%	Up to 10%	34 MPa	-	[80]
Waste brick	5-20%	Up to 10%	38-45 MPa	7.2-7.4 MPa	[63]
Indian clays	5- 25%	Up to 25%	12.1-17.5 MPa	-	[66]

Similarly to cement concrete, fly ash [76] and rice husk ash [79] have also been identified as viable substitute cementitious materials for use in cement mortars. Furthermore, studies have concluded that blends of marble powder with granite powder [77] and Ceramic Powder with Silica Fume [78] at 5-15% replacement level with cement yield compressive strengths of 33-62 MPa, which is higher than individually substituted cement mortars. Goyal et al. (2019) [80] investigated the use of textile sludge into cement mortar varying the replacement proportion 5% to 25%. The findings indicated that a 5% replacement proportion was optimal, yielding a compressive strength of 44 MPa and a tensile strength of 3.8 MPa. Similarly, Chang et al. (2010) [53] investigated the use of sewage sludge ash in cement mortar considering the replacement proportions 10% to 30% and concluded that replacing up to 10% provides the maximum compressive strength of 34 MPa.

2.3 WATER TREATMENT SLUDGE AS CEMENTITIOUS MATERIALS

The hydration reactions of the silicate phases, C3S and C2S, in ordinary Portland cement (OPC) are fundamental to the development of strength in concrete. When C3S reacts with water, it forms calcium silicate hydrate (C-S-H), which is the main strength-giving compound, and calcium hydroxide (CH), which increases alkalinity. Similarly, the hydration of C2S also produces C-S-H and CH, but at a slower rate, contributing to long-term strength. The C3S reaction dominates early strength development, while C2S ensures durability over time. These reactions highlight the importance of silicate phases in achieving the desired mechanical properties of cement. The main reactions involve the silicate phases (C_2S and C_3S) in OPC and WTS with water as shown follows:

$$2C_3S+6H_2O \rightarrow 3CaO \cdot 2SiO_2 \cdot 3H_2O\ (C-S-H) + 3Ca(OH)_2\ (CH)$$

$$2C_2S+4H_2O \rightarrow 3CaO \cdot 2SiO_2 \cdot 3H_2O\ (C-S-H) + Ca(OH)_2\ (CH)$$

Table 2.3 summarizes studies on WTS applied in construction as a partial replacement of cement. Vasudevan (2019) [81] found that WTS could be used to replace up to 10% of cement in concrete, achieving a compressive strength of 36.41 MPa. Similarly, Huang and Wang (2013) [82] found that WTS can replace 40% of aggregate, producing structural lightweight aggregate concrete (726 kg/m^3) and non-structural lightweight aggregate concrete (518 kg/m^3) with compressive strengths ranging from 24.8 to 43.2 MPa. A related study by Sales et al. (2011) [26] demonstrated that aggregate in lightweight concrete for non-structural applications (1847 kg/m^3) can be replaced with a mixture of WTS and sawdust, resulting in compressive and tensile strengths of 11.1 MPa and 1.2 MPa, respectively. Limited research has explored the partial replacement of cement with WTS in cement mortar. Silva et al. (2022) [83] reported that replacing up to 20% of cement with WTS in Self-Compacting Mortar (SCM) achieves a compressive strength of 39 MPa at 28 days. Similarly, Li et al. (2021) [84] studied mortar, replacing up to 15% of cement, fly ash, and sand with WTS, resulting in a compressive strength of 43.8 MPa at 28 days.

Table 2.3. Studies focusing on the use of WTS as alternative materials in construction applications.

Products	Location of Study conducted	Partial re-placement materials	Proportion of replacement	Recommended proportion	Strength at 28 days with recommended proportion	References
SCM	Brazil	Cement	0-20%	10%	39 MPa	[83]
Mortar	Australia	Cement, Fly Ash & Sand	0-15%	5%	43.8 MPa	[84]
Concrete	Malaysia	Cement	5-20%	10%	36.41 MPa	[81]
SCC	Malaysia	Cement	0-20%	15%	70.2 MPa at 180 days	[85]
Brick	China	Clay	0-40%	20%	15.7 MPa at 1200 ºC	[86]
Concrete	Taiwan	Aggregate	40-65%	40%	24.8-43.2 MPa	[82]
Ceramics	Lithuania	Clay	0-40%	5%	48.5-53.8 MPa	[87]
Concrete	Brazil	Aggregate (WTS mixed with saw dust)	n/a	n/a	11.1 MPa	[26]
Brick	UK	Clay (WTS mixed with rice husk)	80-100%	85%	53 MPa at 1100 ºC	[88]
Brick	Taiwan	Clay (WTS Mixed with excavation waste soil)	0-30%	15%	98 MPa at 1100 ºC	[89]

Note: SCM: Self-Compacting Mortar, SCC: Self-Compacting Concrete

There have been few studies conducted on the characterization and use of WTS as substitute materials in construction applications other than concrete and mortar. O'Kelly (2008) [90] investigated the geotechnical properties of WTS and reported that the WTS fall in high plasticity clay (as per USGS classification) (PI=250%, Sp. Gr.=1.86, bulk density 1.04 t/m^3, dry density 0.14 t/m^3) with high plasticity and compressibility and low permeability, which causes high affinity to the water and thus make it useful as binding materials as construction materials [18]. Breesem et al. (2016) [85] found that WTS is suitable for use in Self-Compacting Concrete (SCC) at a maximum dosage of 15%

It also acts as a pozzolanic material, helping to mitigate the effects of magnesium sulfate attack. Moreover, WTS has been also found suitable for use in clay brick. Zhao et al. (2016) [86] reported that up to 20% of clay can be replaced with WTS in brick manufacturing, achieving a maximum compressive strength of 15.7 MPa at 1200ºC. WTS mixed with other materials, such as excavation waste soil and rice husk, is also effective for clay brick production at 1100ºC, resulting in compressive strengths of 53-98 MPa [88,89]. Furthermore, Kizinievič et al. (2013) [87] found that replacing 5% of cement with WTS in ceramics production yields compressive strengths of 48.5-53.8 MPa and densities of 1807-1950 kg/m^3.

Table 2.4 provides an extensive comparison of the chemical composition of substitute cementitious materials and various types of (OPC, offering insights into their potential for use in sustainable construction. Each entry details the percentage composition of major oxides including CaO, SiO_2, Al_2O_3, Fe_2O_3, MgO, TiO_2, Na_2O, K_2O, SO_3, P_2O_5, and loss on ignition (LOI), offering insights into their suitability as supplementary materials in cement production. The SiO_2, Al_2O_3, and Fe_2O_3, contents in pozzolanic substitute materials average approximately 40.1±23.1%, 17.3±14.6%, and 12.0±16.3%, respectively. The substitute materials exhibit diverse chemical characteristics. For instance, fly ash and waste brick are rich in SiO_2, while marble residue has an exceptionally high CaO content. Glass powder combines high SiO_2 with moderate

Na_2O, making it distinctive. Alum sludge shows a high concentration of Al_2O_3, making it a potential alternative for cementitious applications. Similarly, waste textile sludge (WTS) and sewage sludge ash have varied chemical profiles, indicating their adaptability in specialized applications. The OPC types show relatively consistent CaO dominance but vary in secondary components like Al_2O_3 and SiO_2, tailored to meet specific standards (e.g., ASTM and CEM classifications). The LOI values provide additional insights into the thermal stability and purity of these materials.

The hydration reactions of the silicate phases, C_3S and C_2S, in OPC and SCMs are fundamental to the development of strength in concrete. When C_3S reacts with water, it forms calcium silicate hydrate (C-S-H), which is the main strength-giving compound, and calcium hydroxide (CH), which increases alkalinity. Similarly, the hydration of C_2S also produces C-S-H and CH, but at a slower rate, contributing to long-term strength. The C_3S reaction dominates early strength development, while C_2S ensures durability over time. These reactions highlight the importance of silicate phases in achieving the desired mechanical properties of cement. The main reactions involve the silicate phases (C_2S and C_3S) in OPC and WTS with water as shown follows:

$$2C_3S+6H_2O \rightarrow 3CaO \cdot 2SiO_2 \cdot 3H_2O\ (C-S-H) + 3Ca(OH)_2\ (CH)$$

$$2C_2S+4H_2O \rightarrow 3CaO \cdot 2SiO_2 \cdot 3H_2O\ (C-S-H) + Ca(OH)_2\ (CH)$$

The hydration reactions of C3S and C2S are vital for the strength and durability of OPC, producing C-S-H as the primary strength-giving compound. Integrating substitute cementitious materials with diverse chemical compositions offers sustainable alternatives without compromising performance of partially replaced concrete.

Table 2.4. Studies focusing on chemical composition of substitute cementitious materials and different types OPC.

Materials	CaO	SiO_2	Al_2O_3	Fe_2O_3	MgO	TiO_2	Na_2O	K_2O	SO_3	P_2O_5	LOI	References
Unit	%	%	%	%	%	%	%	%	%	%	%	-
Fly Ash	2.67	51.15	20.52	15.73	4.00	1.65	-	2.16	1.28	0.65	-	[91]
Marble residue	77.57	2.72	0.11	0.30	19.11	-	-	0.02	0.08	-	45.31	[92]
Glass powder	9.08	73.64	1.42	0.38	1.43	0.05	11.62	0.01	0.27	0.01	2.09	[67]
Textile sludge	33.5	3.8	0.3	18.9	1.00	-	0.06	0.04	-	-	40.6	[53]
WTS	4.39	52.78	14.38	5.20	3.08	0.62	0.97	3.62	-	0.17	-	[15]
Alum Sludge	0.22	48.82	39.42	5.30	0.44	-	0.08	2.17	0.09	-	-	[85]
Fly Ash	4.85	51.28	24.37	7.92	2.05	-	0.56	2.05	1.53	-	-	
Clay brick waste	0.21	66.72	21.21	6.22	0.48	1.37	0.85		-	0.09	-	[65]
WTS	3.10	54.10	28.84	9.92	0.64	1.28	0.30	0.75	0.54	-	-	[23]
Alum Sludge	1.31	9.41	51.01	0.70	0.22	0.05	0.07	0.20	-	0.32	36.19	[93]
WTS	2.08	63.38	21.50	8.91	2.55	-	0.32	1.14	0.16	-	9.32	[82]
Slag	0.15	42.84	41.20	9.19	3.44	-	0.70	2.12	0.20	-	0.10	[94]
Sewage sludge	1.00	36.5	8.9	3.9	0.60	-	0.40	0.90	4.80	2.50	-	[95]
Iron Slag	4.2	31.50	10.10	56.1	2.00	-	0.20	0.10	1.40	-	-	
Oxygen Furnace	6.90	11.20	0.10	60.70	1.70	-	0.40	0.10	0.20	-	-	
WTS	0.80	49.20	26.30	6.60	1.00	-	0.60	3.20	0.10	-	-	
Marble sludge	58.10	0.80	0.10	0.20	0.80	-	-	0.10	0.10	-	-	
Shale powder	1.35	52.18	14.69	6.23	1.33	-	0.24	0.89	-	-	-	[27]
WTS	1.68	52.18	19.90	6.29	1.38	-	0.97	2.90	-	-	-	
Sewage sludge ash	30.49	20.10	4.47	17.20	12.98	1.90	1.57	0.06	1.83	0.25	-	[80]
Waste Brick	6.06	66.52	14.20	5.45	2.35	-	0.67	2.09	0.75	-	-	[63]
OPC (CEM-I 52.5R)	63.55	20.01	3.03	3.87	4.02	0.58	-	-	2.46	0.21	-	[91]
OPC (CEM-I 42.5 N)	63.55	23.83	3.59	3.87	2.10	0.46	-	0.38	1.71	0.43	-	
OPC	63.10	22.30	5.93	2.80	0.80	-	0.80	0.40	1.30	-	2.5	[53]
OPC (Type-I, ASTM C150)	65.12	20.09	5.60	3.44	1.53		0.09	0.39	2.96	-	2.05	[85]
OPC (CP II–Z–32)	52.79	22.41	6.77	3.15	4.15	-	0.78		2.79	-	-	[65]
OPC (CEM-I 42.5 N)	64.18	21.02	5.04	2.85	7.10	-	0.24	0.70	2.58	-	-	[96]
OPC	64.21	21.5	5.07	4.39	2.00	-	0.23	0.29	3.25	-	2.40	[23]
OPC (CEM-I)	60.94	21.34	5.00	3.38	1.91	-	0.09	0.54	2.04	-	-	[63]
OPC (CPV)	41.42	20.49	3.13	0.08	3.15	-	-	0.99	-	-	4.68	[92]
OPC (CEM-I 52.5N)	64.25	20.07	4.86	3.07	0.95	-	0.18	1.00	3.55	-	1.09	[67]

2.4 LEACHING CHARACTERISTICS OF CONCRETE WITH SUPPLEMENTARY CEMENTITIOUS MATERIALS

It is clear that sludges can be used as partial replacement of cement in concrete. This approach has the advantage of using these sludges within the concrete, which helps to manage the waste materials. The leaching characteristics are impacted by the type and particle size distribution of the waste materials, as well as by the modification methods applied to adapt these materials for use as SCMs in concrete [59]. However, since these sludges are by-products and may have potential environmental risks due to their leaching properties, it is crucial to assess their potential to release harmful substances into the surrounding environment [97,98].

Therefore, it is essential to conduct thorough investigations into the leaching behavior of concrete that incorporates sludges as partial cement replacements to ensure that any potential environmental impacts are properly evaluated and managed. **Table 2.5** summarizes the concentrations of heavy metals like Chromium (Cr), Lead (Pb), Nickel (Ni), Copper (Cu), Zinc (Zn), Arsenic (As), Cadmium (Cd), Manganese (Mn), and Mercury (Hg) found in the leachate samples from concrete specimens mixed with various types of sludge, such as textile, sewage, tannery, and water purification sludge.

Sewage sludge consistently exhibited higher concentrations of heavy metals among the different sludge types compared to others, as evidenced in studies by Li, Tsang, et al. (2017) [99], Li et al. (2017) [100] , and Chen and Poon (2017) [101]. Substantially higher concentrations of Ni, Cu, Zn, and Cr, as seen in **Table 2.5**, highlight the potential environmental risks linked to using sewage sludge in concrete as a substitute construction material. The concentrations of Ni were reported to be as high as 0.45 mg/L, while Cu, Zn, and Cr levels reached up to 3.34 mg/L, 13.77 mg/L, and 15.2 mg/L, respectively.

Table 2.5. Heavy metal concentrations in leachate samples from concrete specimens.

Name of the materials	**Cr**	**Pb**	**Ni**	**Cu**	**Zn**	**As**	**Cd**	**Mn**	**Hg**	**References**
Unit	(mg/kg)	(mg/kg)	(mg/kg)	(mg/kg)	(mg/kg)	(mg/kg)	(mg/kg)	(mg/kg)	(mg/kg)	
Textile sludge	0.031	0.037	0.025	0.021	1.520	0.001	0.001	0.072	0.001	[29]
Sewage sludge ash with recycled glass cullet	n.d.	0.036	0.0885	2.46	2.601	0.052	0.0165	-	-	[101]
Sewage sludge	-	0.03	0.32	2.1	12	0.17	0.036	-	-	[99]
Tannery sludge	0.28-0.745	0.094-0.134	n.d.	0.085-0.216	0.181-1.48	0.017-0.067	n.d.	-		[59]
Sewage sludge		0.40	0.40	2.2	13.77	0.30	-	-	-	[100]
Sewage sludge and glass powder	15.2	0.28	0.45	3.34	4.1	0.77	1.05		<0.002	[102]
Sewage sludge	0.01-0.04	0.01-0.03	0.04-0.09	0.06-0.17	1.02-1.27	-	-	-	-	[103]
Water purification sludge	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	n.d.	-	-	[27]
Sewage Sludge Ash	0.0887-0.0968	n.d	0.0331-0.0388	-	-	0.0073-0.0134	0.2672-0.2739	-	-	[104]

n.d.: not detected

On the other hand, textile and tannery sludge showed relatively lower heavy metal concentrations, as found by Kasaw et al. (2021) [29] and Juel, Mizan, and Ahmed (2017) [59]. Although these values are lower compared to sewage sludge, the presence of heavy metals still raises concerns about their long-term environmental effects. The studies by Wang and Yuan (2015) [104] presented a contrasting view, reporting lower concentrations of heavy metals from sewage sludge ash compared to other studies. This discrepancy highlights the variability in heavy metal content depending on the sludge source and treatment. Surprisingly, water purification sludge did not show detectable levels of heavy metals in the leachate, as reported by Chen, Ma, and Dai (2010) [27]. However, Mn and Hg were less frequently investigated, with only in a few studies, such as Kasaw et al. (2021) [29] and Chen and Poon (2017) [102]. The lower focus on these metals may be due to their lesser prevalence or lower environmental risk compared to the other heavy metals.

2.5 SUMMARY

The literature review section explores the use of various waste sludge materials as partial replacements for cement in concrete and mortar, focusing on their potential benefits and mechanical properties. Research has investigated different sludges such as sewage, textile, and paper waste, with replacement levels ranging from 0% to 50%. Findings suggest that these materials can improve the sustainability of concrete by reducing the cement content while maintaining or even enhancing mechanical strengths, with compressive strengths varying widely based on the type and proportion of sludge used. However, the environmental impact of incorporating sludge into concrete, particularly in terms of leaching characteristics, is crucial. Research highlights that while some sludges, like sewage sludge, may release higher concentrations of heavy metals, others, like water purification sludge, have shown minimal environmental risks.

Chapter 3: MATERIALS AND METHODOLOGY

3.1 GENERAL

This Chapter describes the systematic approach taken to investigate the use of water treatment sludge (WTS) in concrete and mortar. It outlines the raw materials and their characterization, including the quality and suitability of OPC and WTS. The methods for preparing concrete and mortar specimens are detailed, along with statistical tests to analyze correlations among variables. It also includes procedures for sample digestion, semi-dynamic tank leaching to assess heavy metal leaching, and data analysis techniques for evaluating leaching behavior. Lastly, the section covers life cycle impact analysis to assess the environmental implications of using WTS.

3.2 RAW MATERIALS

The physical appearance of the raw materials used in this study for preparing cement mortar and concrete is depicted in **Figure 3.1**. Ordinary Portland Cement (OPC) of CEM-I type (42.5N) is used as the binder (**Figure 3.1**a), while WTS is used as a substitute material for cement in both cement mortar and concrete (**Figure 3.1**b). Standard sand (DIN-EN-196-1) is used as the aggregate in cement mortar (**Figure 3.1**c). On ther other hand, Sand (Sylhet sand) collected from northeastern part of Bangladesh is used as fine aggregate (**Figure 3.1**d) in concrete. Coarse aggregate (**Figure 3.1**e) is made up of crushed angular stone sourced from the Chattogram region used as coarse aggregate in concrete.

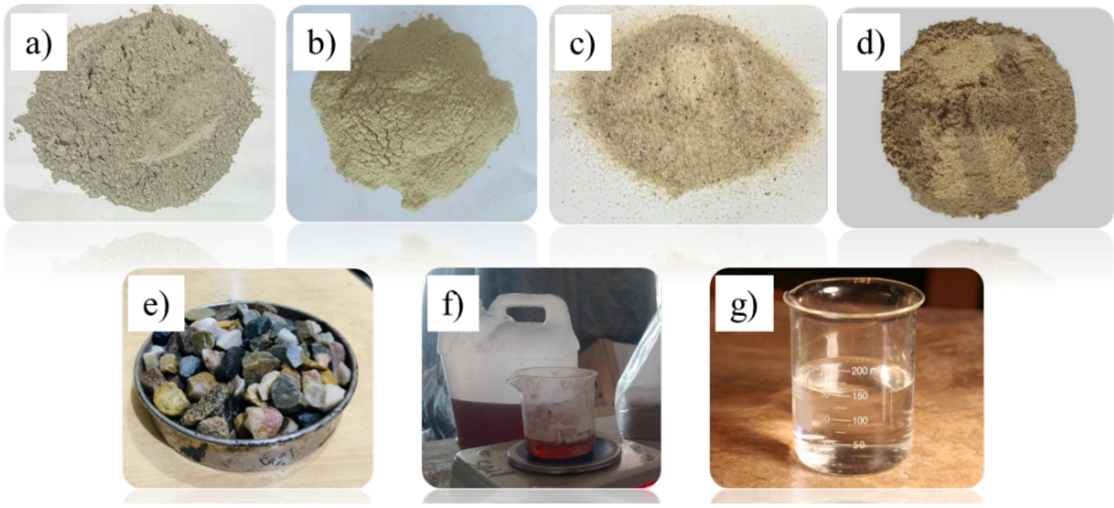


Fig. 3.1 Raw materials used in this study for both cement mortar and concrete (a) CEM-I OPC cement, (b) WTS, (c) DIN-EN-197-1 Standard sand, (d) Sylhet sand, (e) Coarse aggregate, (f) Super plasticizing Admixture and (g) water.

A super-plasticizing admixture (**Figure 3.1**f) is also employed in the concrete. Water that meets drinking quality standards is used during the casting and curing processes of both cement mortar and concrete (**Figure 3.1**g).

3.2.1 Cement

In this study, Ordinary Portland Cement (OPC) (CEM-I) is used to prepare cement mortar and concrete. The physical properties, including specific gravity, normal consistency, setting time, and fineness are listed in **Table 3.1**. The properties were determined using American Society for Testing and Materials (ASTM) standards and Indian Standards (IS), as presented in **Table 3.1**.

Table 3.1. Physical properties of OPC (CEM-I, 42.5N) used in this study

Sl. No.	Name of the Properties		Value	Specifications (BDS EN 197-1:2003)	Test Method	References
1	Specific Gravity of Cement	:	2.91	-	IS:4031	[105]
2	Normal Consistency (%)	:	28	-	ASTM C187	[106]
3	Initial Setting time (minute)	:	82	50 (min)	IS:4031	[107]
4	Final Setting time (minute)	:	310	375 (max)	IS:4031	[107]
5	Fineness (m^2/kg)	:	305	280 (min)	ASTM C430	[108]

The strength class of CEM-I is 42.5N. The primary constituents of this cement type are clinker (95-100%) and gypsum (0-5%). All the properties of the cement comply with the specifications outlined in BDS EN 197-1: 2003 [109].

3.2.2 Water Treatment Sludge (WTS)

WTS was collected from the Sheikh Russel Water Treatment Plant, which is operated by the Chattogram Water and Sewerage Authority (CWASA), Bangladesh. The preparation process for the collected WTS, illustrated in **Figure 3.2**, involved several steps. By the preparation process, the WTS was treated to use as a substitute material in cement mortar and concrete. Initially, the raw sludge was dried in an oven at 105ºC for 24 hours to remove all moisture resulting in dewatered WTS. This dewatered WTS was then placed in a furnace and heated to 500ºC for 4 hours to eliminate any organic substances, including pathogens and other living organisms.

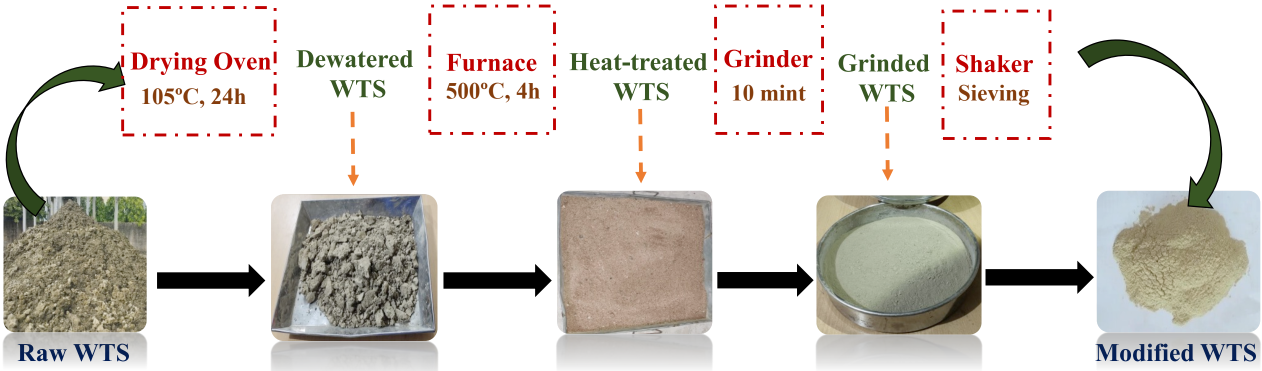


Fig. 3.2 Schematic diagram for the preparation of modified WTS from raw WTS.

Following the heat treatment, the WTS was mechanically ground for 10 minutes using a mechanical grinder. The ground WTS was then sieved through a #200 ASTM sieve (75μm opening) using a mechanical shaker to ensure uniform particle size. The sludge that passed through the #200 sieve was utilized for further investigation in this study. The specific gravity of the WTS was determined as 2.11 following IS:4031 standard.

3.2.3 Fine Aggregate (FA)

In this study, cement mortar was prepared using DIN-EN-196-1 standard sand, while the cement concrete was prepared using locally available sand named Sylhet sand in Bangladesh. The particle size distributions of both types of sands were determined using ASTM C778 standard [110] as shown in **Figure 3.3**. The Fineness Modulus of DIN-EN-196-1 sand and Sylhet sand were calculated as 2.86 and 2.97, respectively. Other physical properties of both sands are given in **Table 3.2**. The test methods followed to determine the properties are also given in the table.

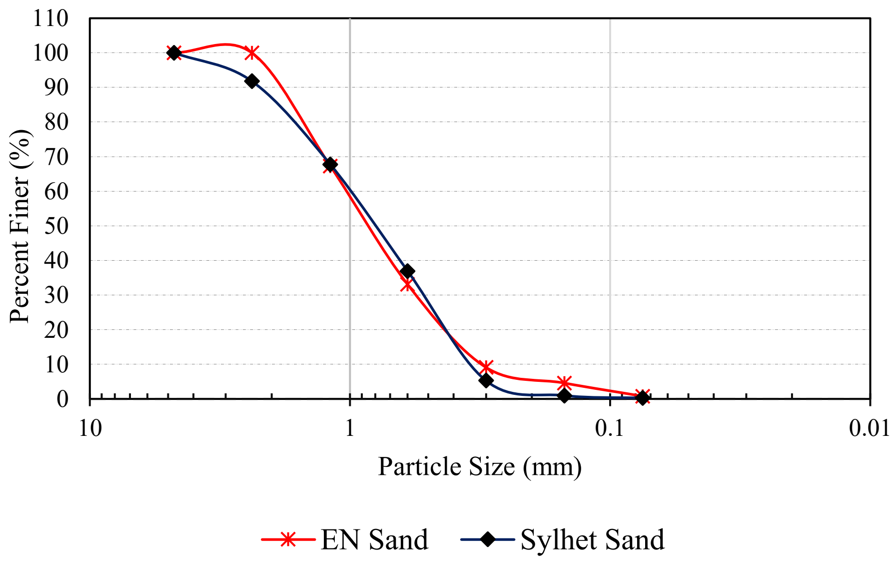


Fig. 3.3 Particle size distribution curves of DIN-EN-196-1 sand and Sylhet sand

Table 3.2. Physical properties of Sylhet sand and DIN-EN-196-1 sand used in this study as FA.

Sl. No.	Name of the Properties		DIN-EN-196-1 sand	Sylhet sand	Test Method	References
1	Fineness Modulus	:	2.86	2.97	ASTM C136	[111]
2	Apparent Specific Gravity (OD basis)	:	2.69	2.54	ASTM C127	[112]
3	Bulk Specific Gravity (OD basis)	:	-	2.34	ASTM C127	[112]
4	Bulk Specific Gravity (SSD)	:	-	2.42	ASTM C127	[112]
5	Dry Rodded Unit Weight (kg/m^3)	:	1586	1648	ASTM C29	[113]
6	Absorption capacity (%)	:	0.83	3.38	ASTM C127	[112]
7	Surface Moisture (%)	:	0.57	1.13	-	-

3.2.4 Coarse Aggregate (CA)

Crushed angular stone chips that satisfies the specifications available in ASTM C33 [114] were used in this study as Coarse Aggregate (CA). Detailed Gradation of the CA with ASTM C33 requirements are given in **Table 3.3**.

Table 3.3. Gradation of CA and ASTM C33 requirements.

Sieve opening	Percent (%) retained	Cumulative Percent (%) retained	Percent (%) Finer	ASTM C33 Requirement (% Finer)
1"	0	0	100	90-100
3/4"	33.12	33.12	66.88	40-85
1/2"	35.65	68.77	31.23	10-40
3/8"	25.62	94.39	5.61	0-15
#4	5.61	100	0	0-5

Similar to fine aggregate, the physical properties of the coarse aggregates were determined using relevant ASTM test methods. The properties of the coarse aggregates are detailed in **Table 3.4**. The results of the physical properties of the coarse aggregates indicate that they have a moderate specific gravity, low absorption capacity, and minimal free moisture content.

Table 3.4. Physical properties of coarse aggregate (CA)

Sl. No.	Name of the Properties		Value	Test Method	References
1	Apparent Specific Gravity (OD basis)	:	2.77	ASTM C127	[112]
2	Bulk Specific Gravity (OD basis)	:	2.67	ASTM C127	[112]
3	Bulk Specific Gravity (SSD)	:	2.72	ASTM C127	[112]
4	Dry Rodded Unit Weight (kg/m^3)	:	1652	ASTM C29	[113]
5	Absorption capacity (%)	:	0.49	ASTM C127	[112]
6	Free Moisture (%)	:	0.28	-	-

3.2.5 Admixture

The application of mineral admixtures such as slag, fly ash, GGBS, WTS etc., reduces the workability of cement concrete. To achieve the workability of the cement concrete, a poly-carboxylic ether-based super-plasticizing admixture is used in this study. It effectively prevents segregation of ingredients, which is particularly beneficial under high temperature conditions where concrete tends to set quickly. The admixture meets the standards available in ASTM C494 code

[115]. The recommended dosage of the admixture is 700 mL per 100 kg of cement with a specific gravity of 1.20.

3.2.6 Water

The water used in this study satisfied drinking water quality [116]. **Table 3.5** provides the technical specifications and experimental values for the instruments used to measure the water quality during curing. The experimental results indicate that the water quality met the required standards [116]. Additionally, the water was free from total coliforms and fecal coliforms, with no detectable levels of iron (Fe), copper (Cu), chromium (Cr), cadmium (Cd), manganese (Mn), and nickel (Ni).

Table 3.5. Technical specifications of the devices/methods used and the experimental values from the laboratory tests for the water used during casting and curing

Parameters	Unit	Name of the Instrument/ Methods	Range	Accuracy	Experimental values
pH	Value	HI9814 (GroLine)	0 to 16.0	± 1.0%	7.4
Color	PCU	HI97727 (Hanna)	0 to 500	± 5.0%	02
Turbidity	NTU	Turbidity Meter-Lutron TU	0 to 1000	± 1.0%	01
TDS	mg/L	HI9814 (GroLine)	0 to 3000	± 2.0%	0.5
EC	mS/cm	HI9814 (GroLine)	0.00 to 6.00	± 2.0%	8.75
Hardness	mg/L	Standard Methods provided by APHA			1.12
TC/FC	count/100 mL	MF USEPA 9132	n/a	± 3.0%	0
Heavy metals	mg/L	Hach DR9000 VIS Spectrophotometer			ND

3.3 CHARACTERIZATION METHODS FOR OPC AND WTS

Using an electronic microscope (JEOL JSM-7610F, operating at 15 KeV with a counting rate of 1096 cps), Scanning Electron Microscopy (SEM) and Energy Dispersive X-ray (EDX) analyses were performed to identify the morphological characteristics and elemental composition (C, O, Na, Mg, Al, Si, S, K, Ca, Fe, Cu, Pt, etc.) of both OPC (CEM-I) and WTS. Images were captured at magnifications ranging from 250 to 50000 times to examine the shape, texture, and particle distribution. ImageJ software was subsequently used to determine the size and surface area of the particles for both OPC cement and WTS. The particle size and surface area distributions for both materials were visualized using Origin Pro v2024 software. In addition, descriptive statistics such as Mean, Median, Coefficient of Variation, Standard Deviation, Minimum, Maximum, Range, Interquartile Range, Skewness, and Kurtosis were calculated for the particle size and surface area of both materials using the Statistical Package for the Social Sciences (SPSS v.23). The chemical compositions, focusing on oxides such as Aluminium Oxide (Al_2O_3), Ferric Oxide (Fe_2O_3), Silicon Dioxide (SiO_2), Sulfur Trioxide (SO_3), Magnesium Oxide (MgO), Sodium Oxide (Na_2O), Potassium Oxide(K_2O), Calcium Oxide (CaO), Titanium dioxide (TiO_2), were determined using X-Ray Fluorescence (XRF) with the Best Detection-Vac28mm method.

The study also used X-ray diffraction (XRD) method to analyze OPC and WTS for their mineralogical crystalline phases and compound compositions. A diffractometer equipped with a monochromatic Cu-Kβ source, curved graphite, and a single-crystal (1D) chromator (operating at 40 kV, 50 mA, 15°/min, with a HyPix-400 horizontal detector) was used. Samples were scanned over a 2θ range of 5°–90° at a scan rate of 1/2° per minute with a 0.001° increment. The mineralogical analysis revealed the primary phases of Di-calcium Silicate (C_2S) — $2CaO{\cdot}SiO_2$ (PDF # 00-033-0302), Tri-calcium Silicate (C_3S) — $3CaO{\cdot}SiO_2$ (PDF # 00-003-0807), Tri-calcium Aluminate (C_3A) — $3CaO{\cdot}Al_2O_3$ (PDF # 04-008-8069), and Tetra-calcium Alumino-ferrite (C_4AF) — $4CaO{\cdot}Al_2O_3{\cdot}Fe_2O_3$ (PDF # 01-074-0803). The typical ranges of C_2S, C_3S, C_3A, and C_4AF in OPC are 10-25%, 50-65%, 9-12%, and 7-11%, respectively [117]. The chemical analysis of OPC cement and WTS was conducted in accordance with the standard test methods outlined in ASTM C144 [118] and details of SEM, EDX, XRF, and XRD tests are available in [67,91–93,119].

3.4 QUALITY AND SUITABILITY ASSESSMENT OF OPC AND WTS

The quality and hence suitability of a binder is assessed in term of some controlling parameters such as Lime Saturation ratio (LSR), Silica Ratio (SR) and Alumina ratio (AR) [27,119,120]. The parameters were determined using **Eqs. 3.1-3.3.** The recommended values for LSR, SR, and AR are 0.80–0.96, 1.7–2.7, and 0.90–1.9, respectively [27,121,122].

$$LSR = \frac{CaO}{2.8\times SiO_2+1.18\times Al_2O_3+0.65\times Fe_2O_3} \quad (3.1)$$

$$SR = \frac{SiO_2}{Al_2O_3+Fe_2O_3} \quad (3.2)$$

$$AR = \frac{Al_2O_3}{Fe_2O_3} \quad (3.3)$$

In addition to the above-mentioned parameters, the Bangladesh Standards and Testing Institution (BSTI) suggests some other parameters to assess the suitability of OPC. The BSTI suggested parameters are CaO/SiO_2 (≥ 2), $(CaO+MgO)/SiO_2$ (≥ 1), and SO_3 (≤ 4.0%) (BDS 197-1:2003 of [109]. Moreover, standards for OPC as specified by BSTI should primarily consist of essential reactive oxides, including SiO_2 (≥25%) and a significant amount of Al_2O_3, with the remainder composed of Fe_2O_3 and other oxides. The presence of reactive CaO for hardening purposes should not exceed by 10%, with free CaO limited to 2.5% [109]. Furthermore, according to ASTM C618-19 [123], a residue must meet the following criteria to qualify as a pozzolanic material: the combined content of SiO_2, Al_2O_3, and Fe_2O_3 must be ≥ 70%, SO_3 content must be ≤ 4%, loss on ignition must be ≤ 10%, and no more than 34% of the material should be retained on a 45 μm sieve. The chemical compositions of WTS were also compared with EN 450-1 [124].

3.5 PREPARATION OF CEMENT MORTAR SPECIMEN

Cement mortar was prepared (**Figure 3.4**) considering arbitrary but commonly used ratios of cementitious material to sand (Sylhet sand) of 1:2, 1:4, and 1:6 to investigate the effect of WTS on compressive strength. To investigate the effect of WTS on the flexural strength of mortar, the ratio of 1:4 was considered, where the Sylhet sand was replaced with EN standard sand (EN 196-1). For each ratio, the OPC was partially replaced by six WTS replacement levels (i.e., 0%, 2.5%, 5%, 7.5%, 10%, and 15%). The mortar with 0% replacement level was treated as control mortar. For compressive strength determination, 50 mm cubical specimens were made according to ASTM C109-16 [125], whereas the specimen dimensions were 40mm×40mm×160mm for flexural strength determination according to BS EN 1015:2019 (BS 2019). **Figure 3.4** illustrates the steps involved in the preparation and testing of cement mortar.

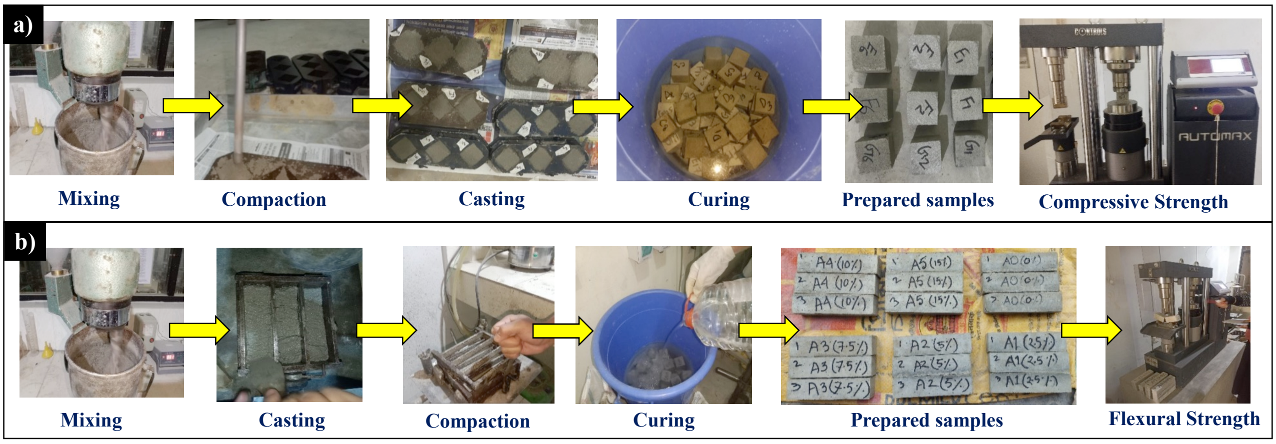


Fig. 3.4 Flow diagram showing the cement mortar specimen preparation for conducting, (a) compressive strength, (b) flexural strength testing.

The specimens were kept in molds for 24 hours in a humid environment before being demolded. The mortar specimens were then cured in water until the testing date. In addition to strengths, the unit weight of the mortar specimens prepared for compressive strength determination were measured following ASTM C138 [126].

3.6 PREPARATION OF CONCRETE SPECIMEN

Similar to cement mortar, two arbitrary but commonly used ratios, such as 1:2:4 and 1:1.5:3, were selected to make concrete specimens to investigate the performances of WTS on cement concrete. The cement content was partially replaced by modified WTS with five different replacement levels of 2.5%, 5%, 7.5%, 10% & 15%). The concrete specimen with no WTS (i.e., 0%) is termed as controlled specimens.

Figure 3.5 illustrates the procedure followed for the preparation of concrete specimens and their subsequent testing. The concrete material was prepared following ACI 211.1 [127]. The slump tests were performed to ensure the workability of the concrete material. For the investigation of compressive strengths, 100 mm cubical specimens and Ø 150 mm × 300 mm cylindrical specimens were made. The specimens were kept in molds in room temperature for 24 hours before curing. The test specimens were immersed in clean fresh water as mentioned earlier for different curing periods. Five curing periods (7, 14, 28 and 72 days) were selected for the series of tests.

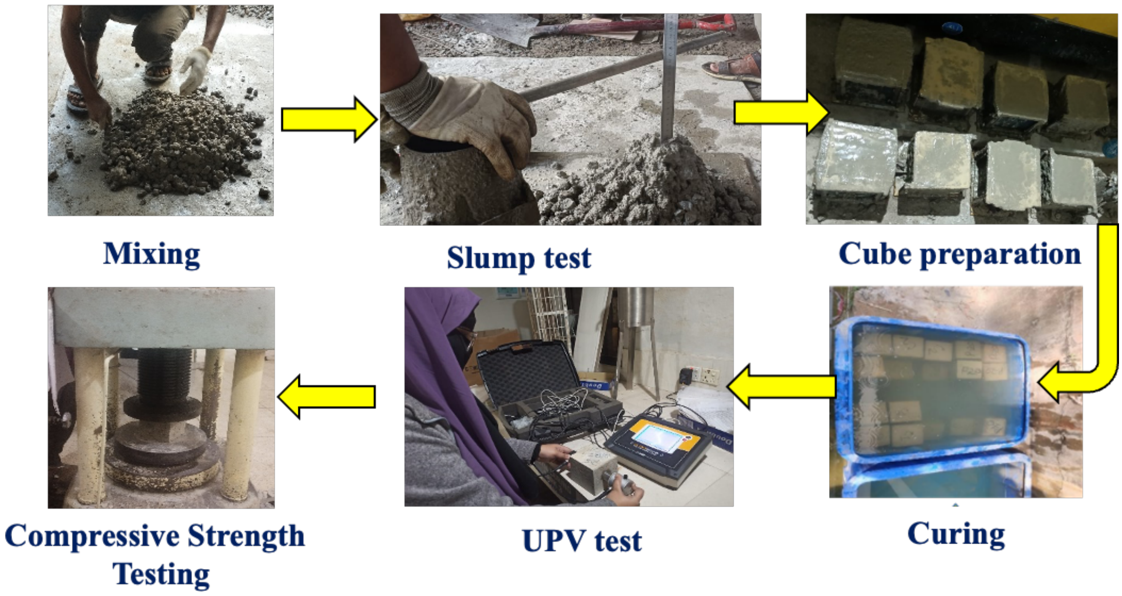


Fig. 3.5 Schematic flow diagram showing the concrete specimen preparation procedure and their subsequent testing.

At the very beginning, the unit weight of concrete specimens were assessed following ASTM C138 code [126] for various mixes with partial replacement of cement by WTS to analyze the trend in weight changes. Ultrasonic Pulse Velocity

(UPV) tests were carried out, following the ASTM C597 guideline [128], on the concrete specimens at the specified curing period to understand the internal cracks, other defects, and level of deterioration of the specimens. The UPV results indicate a range of 3.58 km/s to 4.55 km/s, showing a good quality and uniformity of the concrete samples. The average UPV of 4.09±0.23 km/s suggests that the concrete possesses satisfactory strength and density characteristics [128]. However, a low standard deviation demonstrates minimal variability among the samples, which indicates consistent material properties. The compressive strength tests were than carried out following ASTM C39. Moreover, SEM and XRF tests were also conducted using the methods as mentioned earlier on the concrete specimen with the optimum WTS proportion to gain insights into its microstructure, chemical, and elemental composition.

3.7 STATISTICAL TEST METHODS FOR DETERMINATION OF CORRELATION AMONG CONCRETE VARIABLES

Spearman's non-parametric rank correlation method [129] at a 95% confidence interval was used to determine the association among compressive strength, unit weight, slump value, and the proportion of different materials used in preparing concrete and mortar specimens. This analysis was considered to identify the critical influencing factors, particularly the impact of WTS replacement on the performance of concrete and mortar. Spearman's rank correlation coefficient, r_s, measures the strength of monotonic relationships between variables, accommodating non-normally distributed data. The coefficient is calculated using Eq. (3.4) [129].

$$r_s = 1 - 6\frac{\sum {d_i}^2}{n(n^2-1)} \tag{3.4}$$

Where, n denotes the number of alternatives, and d_i represents the changes between the ranks of two parameters.

The Principal Component Analysis (PCA), a multivariate statistical method, was also performed to filter raw data sets based on component loadings, identifying clusters of parameters with similar characteristics. PCA transforms observed parameters into new orthogonal variables called principal components (PCs), and clustering parameters of similar nature with minimal factor loadings. A loading value of PCs near -1 or +1 indicates a strong impact, while a value near 0 indicates a weak impact. Both correlation analysis and Principal Component Analysis (PCA) were conducted and represented in Origin Pro 2021b.

3.8 DIGESTION OF THE SAMPLE

Figure 3.6 illustrates the steps involved in the acid digestion process of WTS and sand for heavy metal extraction using the USEPA 3050B method [130]. Initially, concentrated HCl and HNO_3 acids were mixed in a precise 1:3 ratio to prepare Aqua Regia, a highly reactive solution capable of dissolving heavy metals. This prepared solution is then repeatedly combined with a representative 1 gm of previously prepared dry WTS sample, with the mixture being thoroughly stirred using a glass stirrer. The mixture was heated in a 250 mL digestion vessel at 90-95°C using a group heater until the brown fumes stopped forming. It was then allowed to cool to room temperature in an open atmosphere.

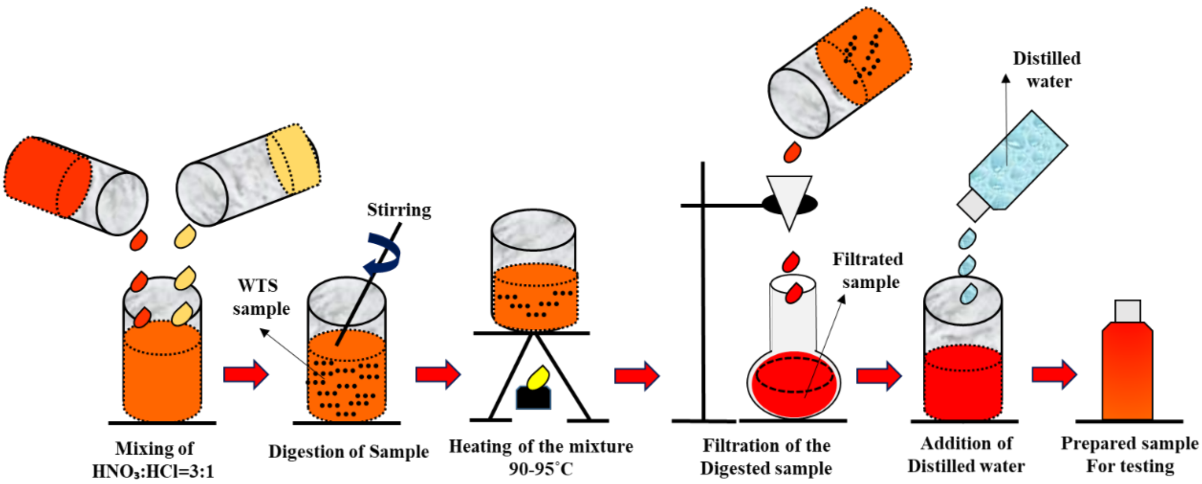


Fig. 3.6 Process involved for the digestion of the WTS sample for the testing of heavy metals.

The cooled mixture was filtered using Whatman No. 41 filter paper. The filter paper and residue were returned to the digestion flask and refluxed with additional concentrated HCl before being filtered again. Finally, the digested sample was diluted with distilled water to a final volume of 100 mL. Then the concentration (mg/L) of the particular parameter of heavy metals was determined in spectrophotometer using Atomic Absorption Spectrometry (AAS) method as in given technical specification in **Table 3.6**. Eq. (**3.5**) is used to convert the concentration from mass per volume (mg/L) to mass per mass (mg/kg).

$$\text{Heavy metal concentration (mg/kg)} = \frac{\text{concentration of metals (mg/L)} \times \text{volume of sample (L)}}{\text{sample weight (kg)}} \quad \textbf{(3.5)}$$

3.9 SEMI-DYNAMIC TANK LEACHING METHOD

The concrete specimens chosen for the leaching study consisted of a control specimen with no replacement (0%) and a specimen with the optimum replacement of cement using WTS. After a curing period of 28 days, the specimens were dried for 24 hours. The leaching tanks were prepared from PVC pipes, each with a diameter of 150 mm and a height of 300 mm. These tanks were meticulously cleaned using distilled water and then left to dry in the open air. Distilled water, having the quality parameters specified in **Table 3.6**, was added to the leaching tanks at a rate of 10 mL per square centimeter. Once the concrete specimens were submerged in the tanks, the tanks were sealed securely with polyethylene sheets that had been cleaned beforehand, ensuring no contamination from external sources.

The USEPA 1315 semi-dynamic leaching test [30] was used (**Figure 3.7**) to detect the concentrations of different heavy meals (Zn, Pb, Cu, Mn, Ni, Cd and Fe). The concrete samples [(0% and 5% WTS replacement) (S_1, S_2, ----, S_n)] was immersed in the leaching tanks and collected leachates (L_1 to L_n) were sampled, drained and replaced with 10 mL/ cm² distilled water for different time intervals ($\Delta t_1, \Delta t_2 \dots \dots \dots \dots \Delta t_n$).

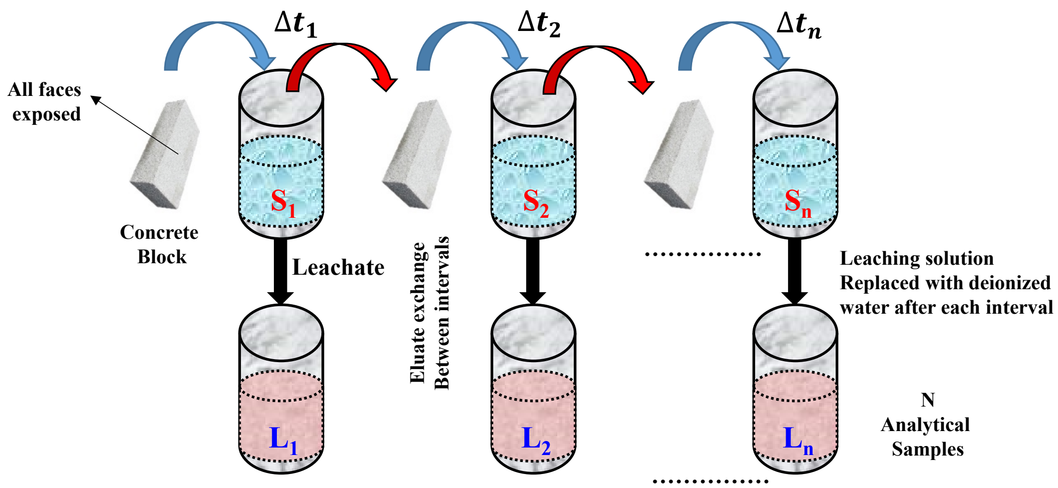


Fig. 3.7 Steps for conducting USEPA 1315 Semi-Dynamic Tank Leaching for n leachate solutions at n leaching period intervals (Δt_1 to Δt_n) (adopter after [28] and [30])

Table 3.6. Technical specification of the devices and methods used in the laboratory tests for heavy metal testing.

Parameters	Name of the Instruments	Methods	Range	Accuracy	Standards[a]
Fe	Hach DR9000 VIS Spectrophotometer	Method 10287	0.001 to 0.10 mg/L	±1%	-
Cu		Method 8143	0.001 to 0.21 mg/L	±1%	100 mg/L
Cr		LCS313	0.002–0.25 mg/L	±5%	05 mg/L
Cd		Method 8192	0.001 - 0.50 mg/L	±1%	01 mg/L
Mn		Method 8149	0.001 to 0.70 mg/L	±5%	260 mg/L
Ni		Method 8150	0.001 to 1.00 mg/L	±5%	1.3 mg/L

[a]Permissible limit set by the United States Environmental Protection Agency (USEPA) standard (Source: [9,30,97,131]).

The leachate samples were collected for 2 h and 1, 2, 7, 14, 28, 42, 49, and 63 days. The collected leachate samples were investigated to measure the concentrations of various heavy metals using the Atomic Absorption Spectrometry (AAS) method. Detailed information regarding the analysis process and the specific heavy metals being assessed is outlined in **Table 3.6.**

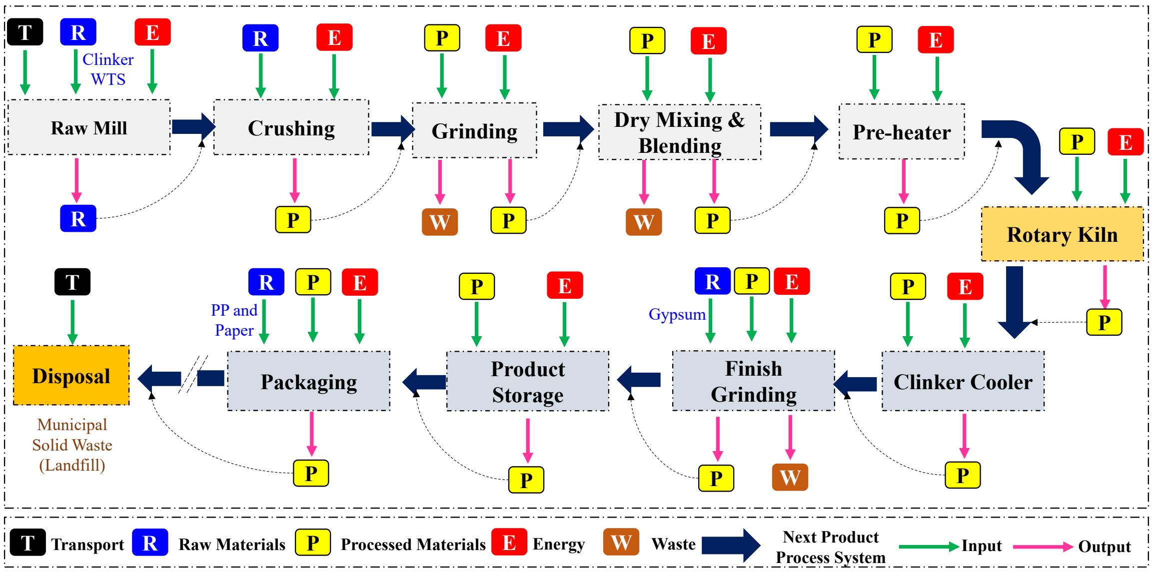
3.10 DATA ANALYSIS AND STATISTICAL METHODS FOR THE INVESTIGATION OF THE LEACHING BEHAVIOR

The compressive strength of two concrete specimens with different levels of WTS replacement is compared with the control specimen. Similarly, the concentrations of heavy metals in the leachate are represented and compared. Moreover, the study examines the trend in leachate parameters over 63 days for both the control specimen and the specimen with 5% WTS replacement. The analysis of the changes in heavy metal concentrations within the leachate during the leaching periods was carried out using the Theil–Sen estimator [132], which is known for its robustness in estimating linear trends. The Mann-Kendall method [133] was also used to determine the statistical significance of these trends. Two statistical methods are essential in identifying whether the observed trends in heavy metal concentrations are significantly positive or negative. Different levels of statistical confidence were used in this study to classify the significance of these trends: 90%, 95%, 99%, and 99.99%, corresponding to p-values of $p<0.10$, $p<0.05$, $p<0.01$, and $p<0.001$, respectively.

These intervals help in distinguishing between trends that are merely suggestive and those that are highly significant. Furthermore, a paired sample T-test (two-tailed) was performed at a 95% confidence interval) using statistical software SPSS (Ver. 29) to investigate the differences in leachate parameters across the various concrete mix ratios. This statistical test was used to assess whether there were significant differences in the leachate characteristics between different mixtures, providing a deeper understanding of how the replacement of cement with WTS affects the leaching behavior. This analysis aims to determine whether the mix ratio or the addition of WTS significantly impacts the concentration of heavy metals in the leachate. The statistical methods are detailed in the studies by Mullick et al. (2019), Pal & Masum (2021), and Ural et al. (2022) [134–136].

3.11 LIFE CYCLE IMPACT ANALYSIS

The life cycle assessment (LCA) was carried out in this study following the guidelines of the DIN EN ISO 14040 [137] method using OpenLCA 2.0 software. The four main stages of Life Cycle Assessment (LCA) include goal and scope definition, Life Cycle Inventory (LCI), Life Cycle Impact Assessment (LCIA), and interpretation of the impact results. These stages are comprehensively discussed by Stafford et al. (2016) [138], Wolde et al. (2024) [139], and Thwe, Khatiwada, and Gasparatos (2021) [140]. **Figure 3.8** illustrates the major inputs and outputs at each stage, defining the system boundaries adopted in this study.



Note: "T" denotes the various transportation methods employed, "R" refers to the raw materials used in cement production, such as clinker and ETS, "P" represents the materials that have been processed at each stage of production, "E" stands for the energy consumed during cement manufacturing, primarily electricity, and "Waste" encompasses all waste materials generated at each step of the production process.

Fig. 3.8 : System boundary of cement production adopted in this study

Primary data was collected from the three representative cement factories in Bangladesh, while secondary data was sourced from the ecoinvent 3.8 UNIT Regionalized database [141] (freely available for low and middle-income countries), specifically for the data of raw material extraction and transportation to the regional port from the sourced country to Bangladesh. Necessary calibration and sensitivity analysis were carried out to verify the accuracy and reliability of the collected data. This study employs the cradle-to-grave approach to assess the environmental impacts associated with cement production. The cement production process aligns with the description provided by Thwe, Khatiwada, and Gasparatos (2021)[140]. The Cradle to Gate phase encompasses activities from raw material extraction and transportation to the delivery of these materials at the raw mill within the cement factory. The Gate to Gate phase covers the cement production process, ends up with the packaging of the cement.

Finally, the Gate to Grave phase considers various applications of the cement, finishing in its disposal. The study assumes that waste is disposed of as municipal solid waste at a nearby landfill, which is s based on common waste management practices in Bangladesh and the typical end-of-life scenario for cement-based materials. The functional unit used for comparison is 1,000 kg of OPC, with two scenarios examined: traditional OPC production and OPC production with a 2.5% to 15% replacement of clinker by WTS. This study used the ReCiPe Midpoint (H) method to assess Eighteen (18) different environmental impacts. These impacts represent the effects of cement production on local, regional, and global scales [140].

3.12 SUMMARY

In this study, cement mortar and concrete were prepared using Ordinary Portland Cement (OPC), water treatment sludge (WTS), and various aggregates. The raw materials included OPC (CEM-I type), WTS, standard sand, Sylhet sand, coarse aggregate, super-plasticizing admixture, and drinking water. Characterization of OPC and WTS involved SEM, EDX, XRF, and XRD analyses to assess their physical and chemical properties. Concrete specimens with various WTS replacement levels were tested for unit weight, compressive strength, and ultrasonic pulse velocity. Cement mortar was prepared and tested for compressive and flexural strengths. Statistical analyses, including Spearman's correlation and Principal Component Analysis, were used to evaluate the effects of WTS. Heavy metal leaching was assessed using the USEPA 1315 method, and life cycle impact analysis was conducted using OpenLCA to compare traditional OPC with OPC containing 2.5-15% WTS.

Chapter 4: RESULTS AND DISCUSSION

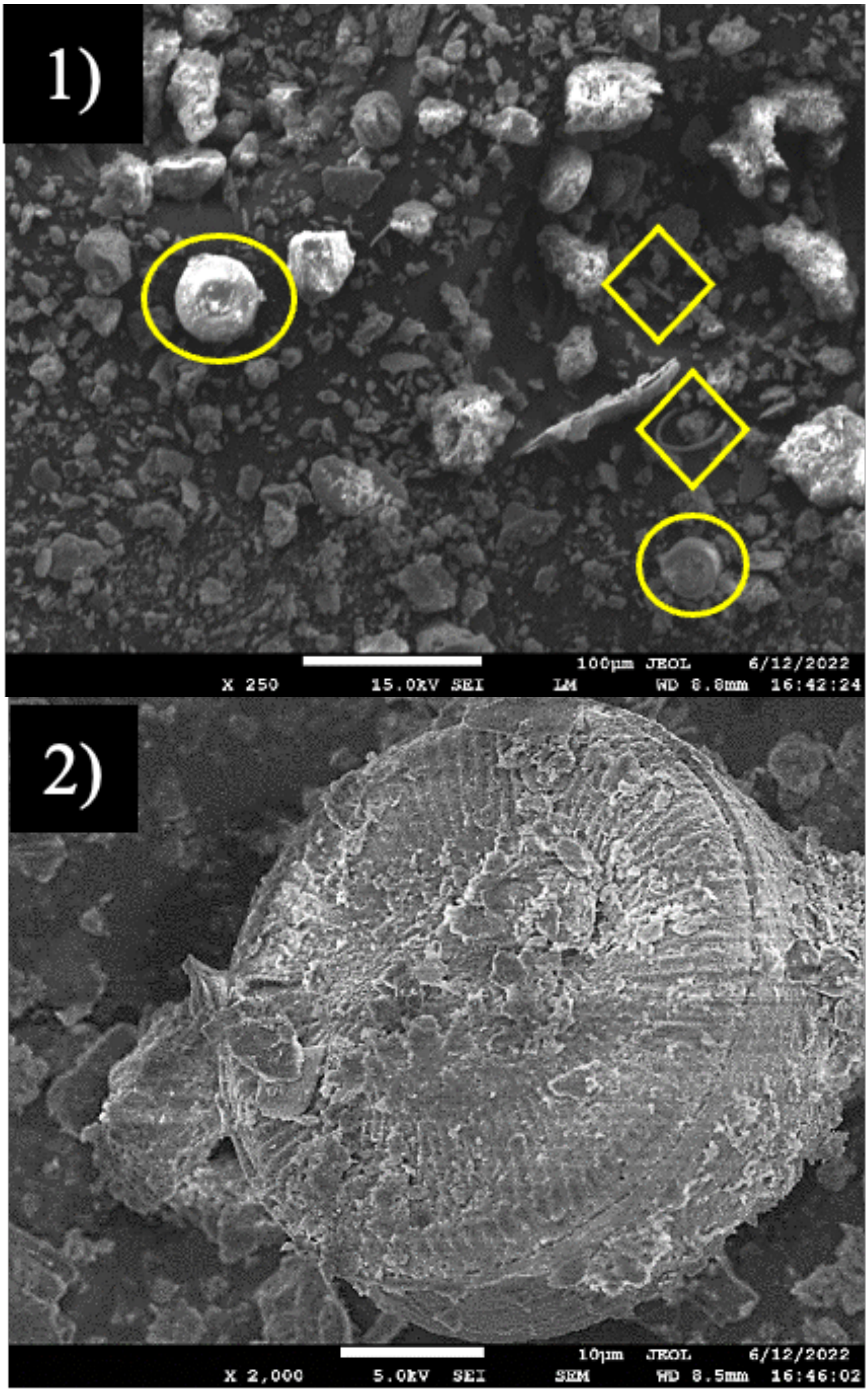
4.1 GENERAL

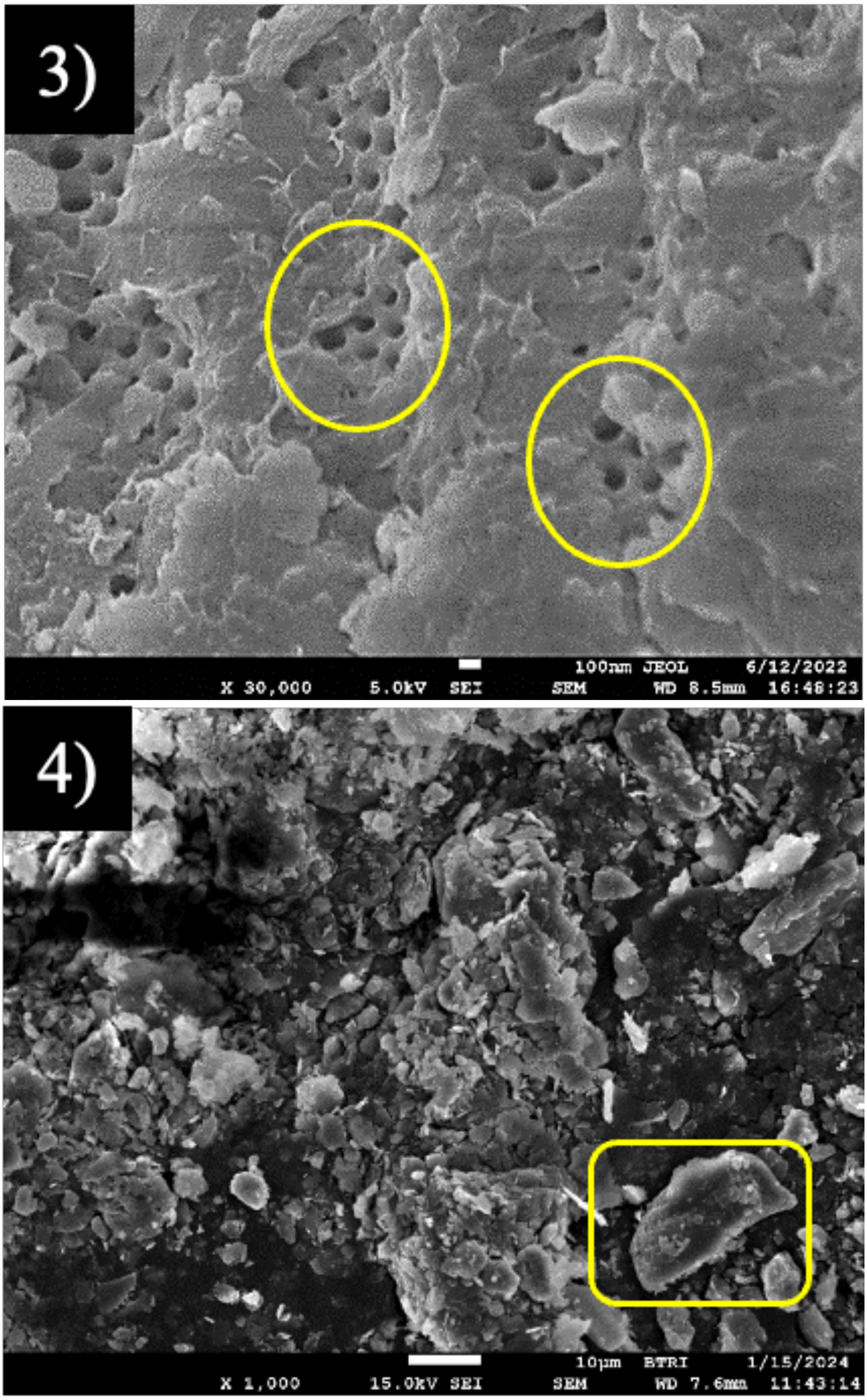
This chapter explores the characterization of binders, focusing on their microstructural, elemental, and chemical properties. It compares Water Treatment Sludge (WTS) and Ordinary Portland Cement (OPC), examining how their distinct characteristics influence concrete performance. The chapter investigates how replacing of OPC with WTS affects concrete's compressive strength, workability, unit weight, and microstructure. It also evaluates the performance of cement mortar containing WTS, assessing variations in compressive and flexural strength, as well as unit weight. Additionally, it addresses the leaching of heavy metals from WTS and sand, analysing changes in concentration and their environmental impact. Finally, the chapter investigates the life cycle environmental impacts associated with cement production, offering insights into the broader implications of using alternative binders in construction.

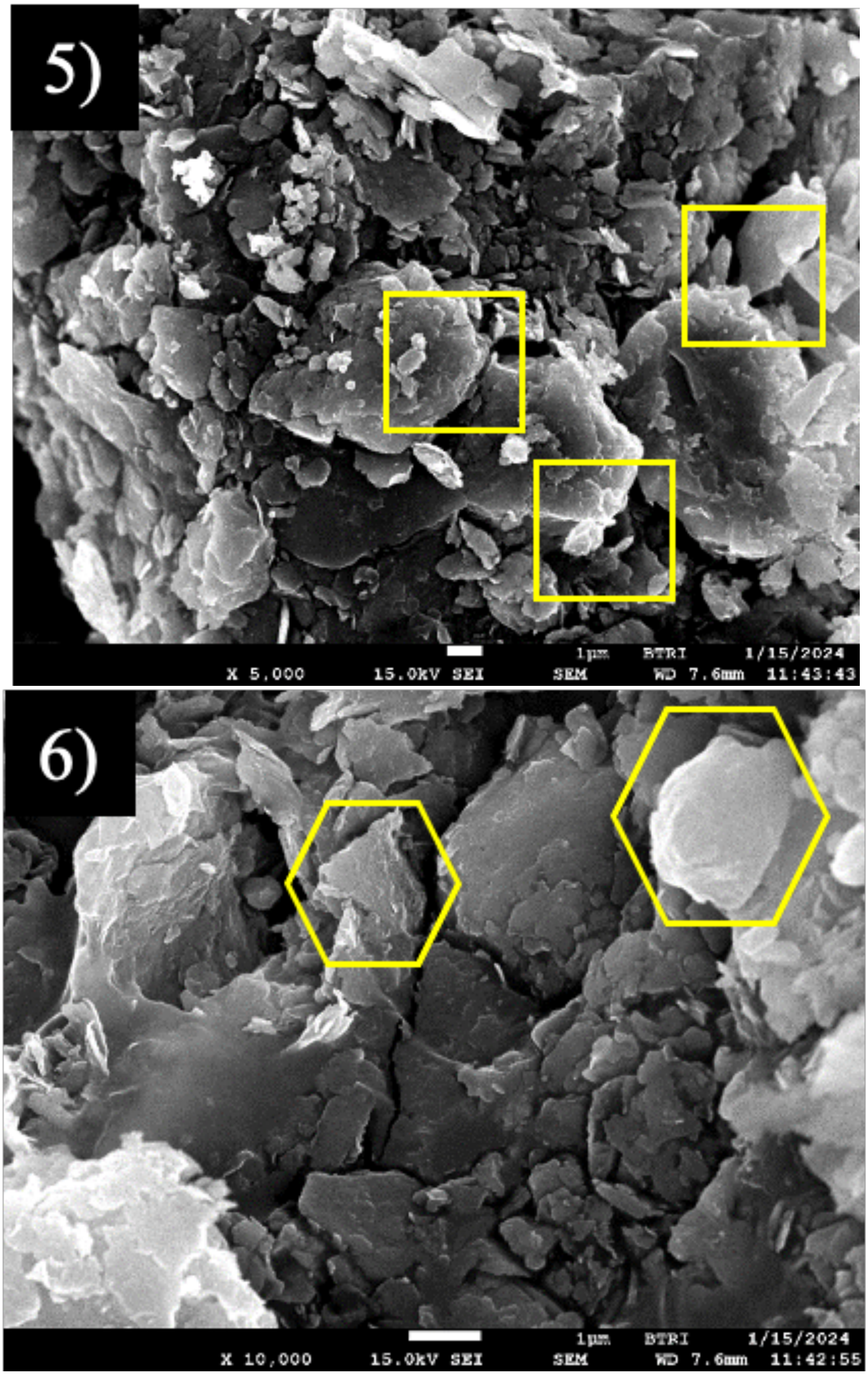
4.2 CHARACTERIZATION OF BINDERS

4.2.1 Microstructural properties of binders

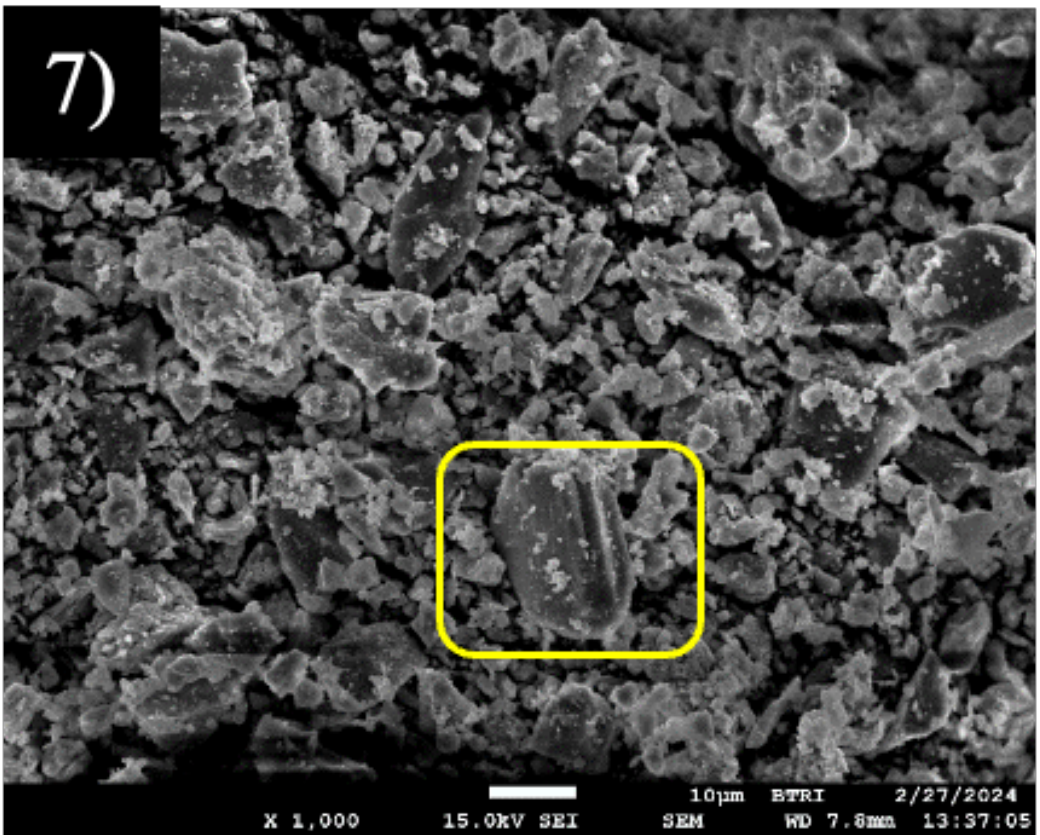
The SEM images presented in **Figure 4.1** show the distinct particle morphologies of both WTS and OPC, highlighting notable differences and similarities in their cementitious characteristics. The particles in both OPC and WTS mainly exhibit irregular shapes, as depicted in the figure. This irregularity enhances the surface area available for hydration reactions and hence potentially boosting the strength and durability of the cement [142,143].

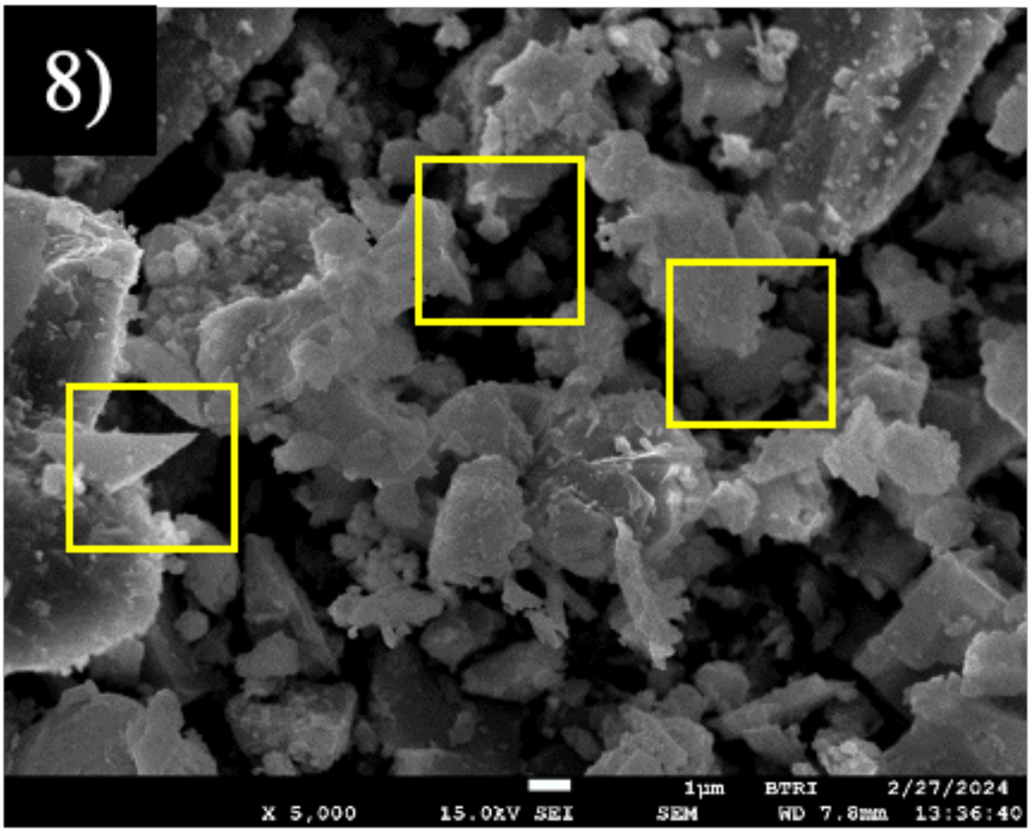

1)
100μm JEOL 6/12/2022
X 250 15.0kV SEI LM WD 8.8mm 16:42:24
2)
10μm JEOL 6/12/2022
X 2,000 5.0kV SEI SEM WD 8.5mm 16:46:02

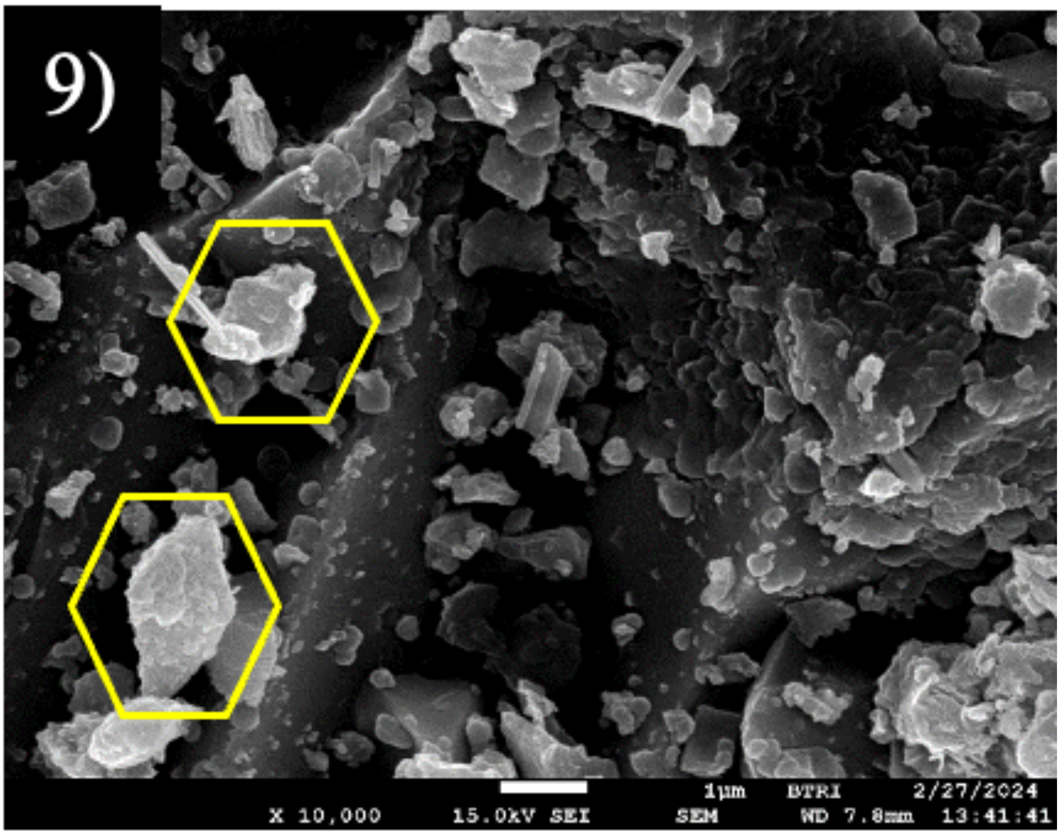

3)
100nm JEOL 6/12/2022
X 30,000 5.0kV SEI SEM WD 8.5mm 16:48:23
4)
10μm BTRI 1/15/2024
X 1,000 15.0kV SEI SEM WD 7.6mm 11:43:14


5)
1µm BTRI 1/15/2024
X 5,000 15.0kV SEI SEM WD 7.6mm 11:43:43
6)
1µm BTRI 1/15/2024
X 10,000 15.0kV SEI SEM WD 7.6mm 11:42:55

(a)


7)
10μm BTRI 2/27/2024
X 1,000 15.0kV SEI SEM WD 7.8mm 13:37:05


8)
1μm BTRI 2/27/2024
X 5,000 15.0kV SEI SEM WD 7.8mm 13:36:40



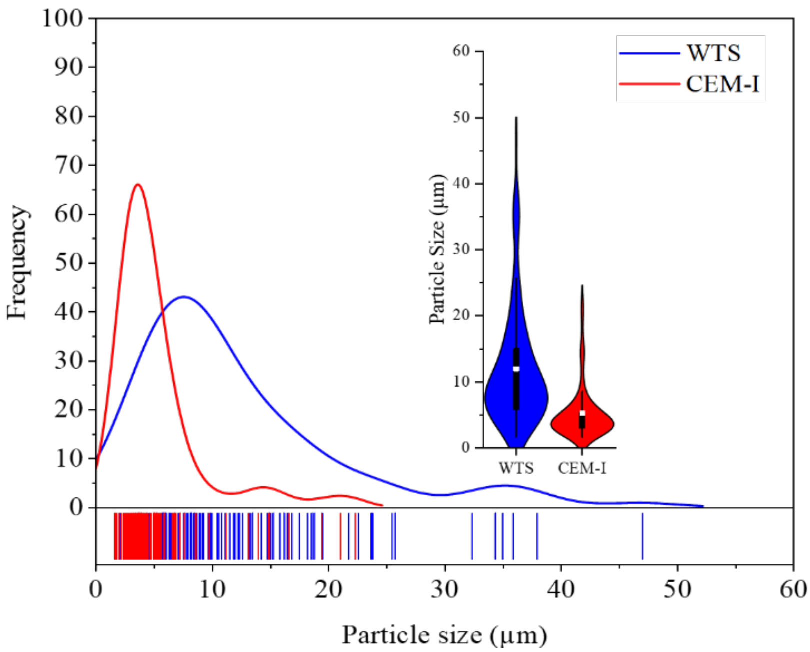
(b)

Fig. 4.1 : SEM images of (a) WTS and (b) OPC/CEM-I cement at different magnifications

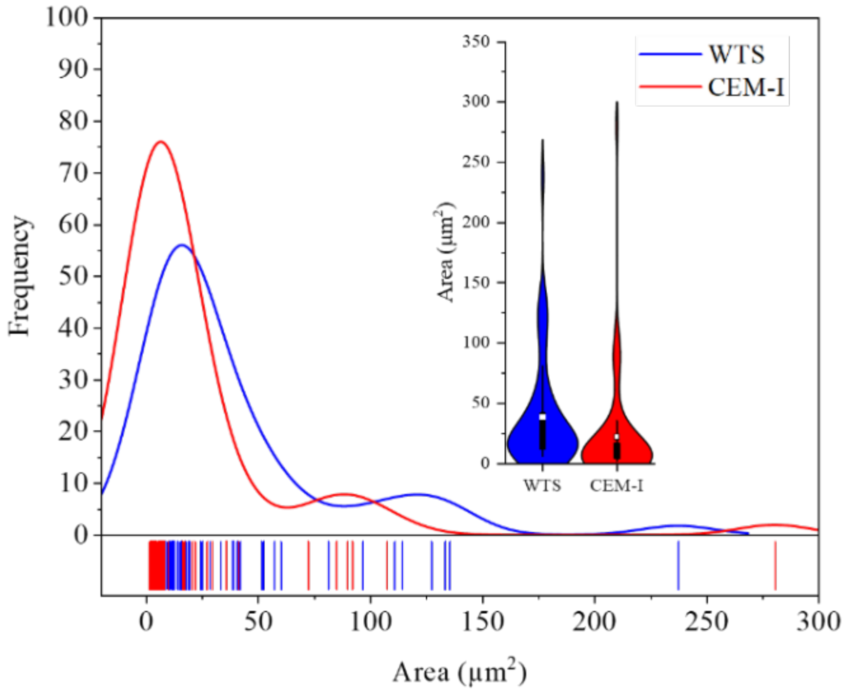
The sharper edges observed in OPC particles (**Figure 4.1** b(8)) and WTS particles (**Figure 4.1** a(6)) indicate high reactivity, promoting efficient interaction with water molecules during the hydration process [144,145]. The increased surface roughness and sharpness aid in mechanical interlocking within the cement paste, thereby enhancing the mechanical properties of the hardened cement paste. WTS particles, while predominantly irregular in shape as depicted in **Figure 4.1** (4, 5), also include a small number of semi-circular and circular particles with diameters ranging from 2.81 μm to 4.81 μm (**Figure 4.1** a(1, 2)). A very small portion of WTS particles are found fibrous (**Figure 4.1** a(1)). The blunter edges of a small portion of WTS particles (**Figure 4.1** a(5, 6)) suggest lower reactivity compared to OPC cement, potentially affecting the rate and extent of hydration [146,147]. Additionally, the presence of pores within the circular or semi-circular WTS particles, with cross-sectional areas ranging from 0.0015 μm^2

to 0.012 μm² (**Figure 4.1** a(3)), could facilitate water ingress, aiding in hydration but potentially increasing porosity and hence might reduce mechanical strength.

The distribution of particle size and particle surface area for WTS and OPC/CEM-I are shown, using Kernel smooth curve with Scott bandwidth, in **Figure 4.2**, where **Figure 4.2(a)** shows the particle size distribution and **Figure 4.2(b)** shows the particle surface distribution. The experimental analysis reveals that WTS particles have an average size of 11.99 μm, compared to 5.23 μm for OPC/CEM-I. The particle sizes for WTS at the 25th, 50th, and 75th percentiles are 5.86 μm, 9.20 μm, and 15.10 μm, respectively, whereas for OPC/CEM-I, the particle sizes are 3.01 μm, 3.88 μm, and 5.48 μm. The suggested range of particle size for OPC/CEM-I spans from 1 μm to 100 μm [148], which aligns well with WTS. The coefficient of variation (CV) for particle size is 73.57% for WTS and 76.36% for OPC which is consistent with [85]. The WTS exhibits a slightly lower degree of variation in particle size.



(a)



(b)

Fig. 4.2 : Distribution of (a) particle size and (b) surface area of the OPC/CEM-I and WTS

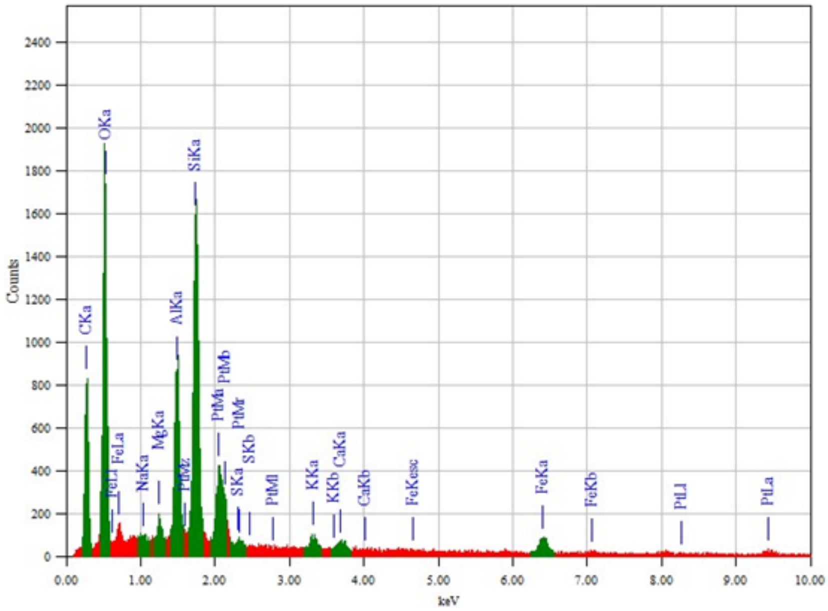
Figure 4.2(a) shows a more uniform and flattened particle size distribution for WTS compared to OPC/CEM-I, suggesting a broader range of particle sizes in WTS. This is consistent with the SEM morphology shown in **Figure 4.1(a)**, which reveals a variety in particle shapes such as irregular, circular, semi-circular, and a few fibrous particles. This broad size range can improve packing density of cement paste by reducing voids in the cement matrix. Both WTS and OPC exhibit almost similar particle surface area distributions as depicted in **Figure 4.2(b)**. The particle surface areas for WTS at 25^{th}, 50^{th}, and 75^{th} percentiles are 12.06 μm^2, 18.91 μm^2, and 42.01 μm^2, respectively and those for OPC are 4.06 μm^2, 5.77 μm^2, and 17.18 μm^2. A larger surface area can facilitate better hydration and stronger bonds within the cement paste, contributing to the overall durability and strength of the cement. The coefficient of variation (CV) for surface area is 117.97% for WTS and 203.73% for OPC, demonstrating substantial variability in both materials. However, WTS exhibits a considerably lower degree of variation in surface area compared to OPC.

4.2.2 Elemental and chemical properties of WTS and OPC

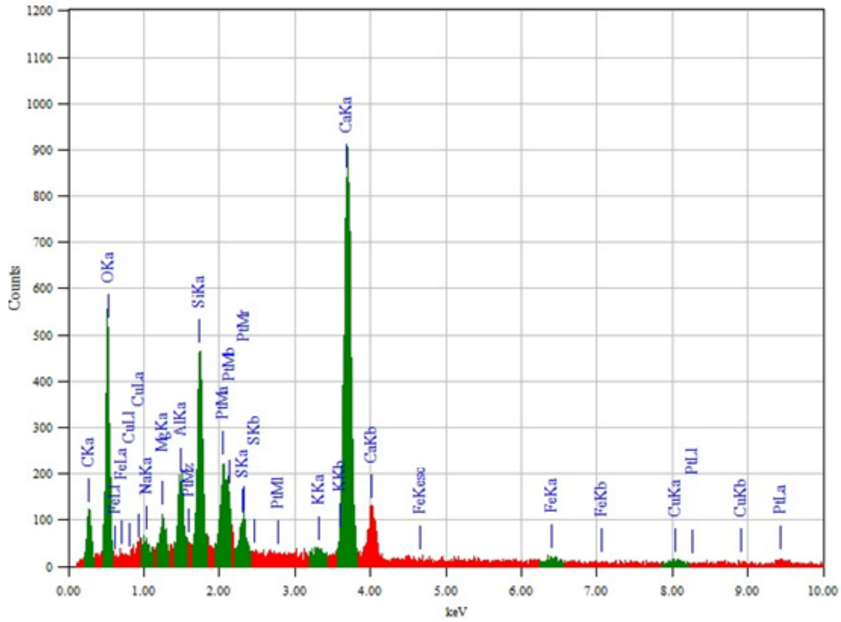
Figure 4.3 displays the representative EDX spectrum with the identified elements found in both WTS and OPC, corresponding to the area at 5000 times magnifications as shown in **Figure 4.1**. The EDX analysis reveals that WTS sludge has a higher Oxygen (O) content compared to OPC cement (see **Table 4.3**). The predominant elements in WTS, ordered by mass percentage, are oxygen (38.76%), carbon (31.81%), silicon (9.10%), and aluminum (4.48%). These elements collectively contribute significantly to the composition, indicating a substantial presence of organic and siliceous components in the sludge. Conversely, in OPC/CEM-I, the composition is characterized by higher proportions of oxygen (36.37%), calcium (29.04%), silicon (5.98%), and aluminum (2.17%) by mass percentage. This higher Oxygen content indicates the presence of various oxides such as SiO_2, CaO, Al_2O_3, Fe_2O_3, and MgO. These oxides enhance the mechanical properties of concrete by creating strong interlocking bonds within the matrix, thereby potentially improving the overall durability and strength of the material [149–151].

The presence of significant Silicon (Si) content in WTS (9.81% by mass) is particularly noteworthy, as silicon plays a major role in strength development through the formation of calcium silicate hydrates (C-S-H) [152–155]. The available silicon in WTS is likely to enhance the mechanical properties due to the influence of C_2S (dicalcium silicate) and C_3S (tricalcium silicate) compounds which are known to significantly impact compressive strength [148,156]. The aluminum (Al) content in WTS, which is almost similar to that in OPC, plays an important role in the early hardening of concrete.

The aluminum facilitates the formation of C_3A (tricalcium aluminate) compounds, which accelerate the initial setting time of the concrete [157]. The higher concentrations of silicon (Si) and aluminum (Al) found in WTS are consistent with those observed in other mineral admixture such as fly ash [158]. The detailed elemental compositions of WTS including carbon, iron and magnesium may supports its potential reactivity and bonding capabilities in cement matrix according to [159]. Calcium, in particular, stands out as a major constituent alongside oxygen, is underscoring its integral role in cement formation [152,153].



(a)



(b)

Fig. 4.3 : EDX spectrum with identified elements: (a) modified WTS and (b) OPC (5000 times magnifications).

Table 4.1. Elements found from EDX analysis for the OPC and WTS

Parameters		C	O	Na	Mg	Al	Si	S	K	Ca	Fe	Cu	Pt
CEM-I	Mass (%)	8.90	36.37	0.25	1.09	2.17	5.98	1.67	0.35	29.04	0.84	2.44	10.91
	Atom (%)	17.39	53.39	0.26	1.05	1.89	5.00	1.22	0.21	17.02	0.35	0.90	1.31
WTS	Mass (%)	31.81	38.76	0.16	0.64	4.48	9.10	0.17	0.81	0.55	3.84	-	9.67
	Atom (%)	46.04	42.11	0.12	0.46	2.88	5.63	0.09	0.36	0.24	1.20	-	0.86

The chemical compositions of WTS and OPC, as analyzed by XRF, are given in **Table 4.2**. WTS exhibits a notable SiO_2 content of 45.40%, exceeding the minimum threshold of 25% proposed by BDS 197-1:2003 for pozzolanic substitute materials [109]. This high silica content indicates a strong potential for WTS to act as a pozzolan, enhancing cementitious properties such as durability and resistance to chemical attack [153,155,160,161]. Additionally, its significant Al_2O_3 and Fe_2O_3 content (18.80% and 18.40%, respectively) also align with standards for effective pozzolanic activity. Furthermore, the combined content of SiO_2, Al_2O_3, and Fe_2O_3 amounts to approximately 81.6%, surpassing the minimum thresholds set by ASTM C618-19 [123].

Table 4.2. Chemical compositions found in WTS and OPC from XRF analysis

Composition		WTS	OPC	Standard Specifications for Natural pozzolanic substitute materials use as cementitious materials		
		Wt (%)	Wt (%)	BDS 197-1:2003 [109]	ASTM C618-19 [123]	EN 450-1 [124]
SiO_2	:	45.40	21.00	≥ 25%	≥70%	≥70%
Al_2O_3	:	18.80	6.83	Significant content		
Fe_2O_3	:	18.40	2.19	Significant content		
K_2O	:	6.15	0.65	-	-	
MgO	:	4.28	2.90	-	-	
CaO	:	1.74	59.00	≤10%	-	≤10%
TiO_2	:	1.70	0.41	-	-	
Na_2O	:	1.43	0.39	-	-	≤ 5%
SO_3	:	0.495	3.93	≤ 4%	≤ 4%	≤ 3%
Other oxides	:	1.44	0.94	-	-	
LOI	:	0.17	1.76	≤ 5%	≤ 10%	≤ 5%

The elevated content of Al_2O_3 and Fe_2O_3 is may be due to the presence of different Aluminum salts such as $Al_2(SO_4)_3.18H_2O$ and Iron salts such as $FeCl_3.6H_2O$, $FeCl_2$, $FeSO_4.7H_2O$ in the WTS those are used as coagulant in water treatment plants [15,26]. Moreover, SiO_2 in WTS may be stemmed from natural geological sources along with potential contributions from industrial discharges and agricultural runoff. The WTS also contains small amounts of minor oxides (CaO, MgO), alkali oxides (K_2O and Na_2O), and additional compounds (TiO_2, P_2O_5, SO_3). Again, WTS is characterized by its CaO content of 1.74%, which adheres to standards specifying ≤10% calcium oxide for natural pozzolans according to BDS 197-1:2003 [109]. In contrast, OPC is characterized by a dominant CaO content of 59.00%, crucial for the hydration process that forms the cementitious matrix and contributes to early strength development, meets BDS 197-1:2003 specifications [109]. WTS exhibits a low LOI of 0.17%, well below the ≤ 5% limit. Moreover, the lower loss on ignition (LOI) of WTS compared to OPC indicates less organic and volatile content, which can positively impact the consistency and stability of the cement mixtures during production and curing stages. SO_3 leads to the instability of cement, resulting in deterioration, swelling, and eventual breakdown of concrete and mortar [162]. WTS contains 0.495% SO_3, comfortably within the allowable limit of ≤ 4%.

Table 4.1 and Table 4.2 provides a detailed comparison of the chemical compositions available in the studied WTS with various substitute cementitious materials and different types of Ordinary Portland Cement (OPC). The SiO_2, Al_2O_3, and Fe_2O_3, contents in pozzolanic substitute materials shows similar values to those of WTS. Notably, WTS exhibits a higher Fe_2O_3 content compared to most pozzolanic substitutes, indicating potential advantages in terms of pozzolanic reactivity and strength enhancement in concrete. The total combined content of SiO_2, Al_2O_3, and Fe_2O_3 in WTS is notably higher than that of other cementitious materials given in **Table 4.1**, indicating its substantial contribution to the overall mineral composition and potential performance benefits in cementitious blends. Moreover, WTS demonstrates a lower LOI content compared to many other materials, including OPC, indicating reduced organic and volatile content that could positively influence concrete production and environmental sustainability than other materials. Similarly, the sulfate (SO_3) content in WTS is comparatively lower, aligning with desirable limits for cement additives and ensuring compatibility with standard specifications.

4.2.3 Mineralogical properties of WTS and OPC

The mineralogical compositions available in WTS and OPC are analyzed using X-ray diffraction (XRD) method shown in **Figure 4.4**. The compositions are checked for the recommended values proposed in the existing literature as presented in **Table 4.3**.

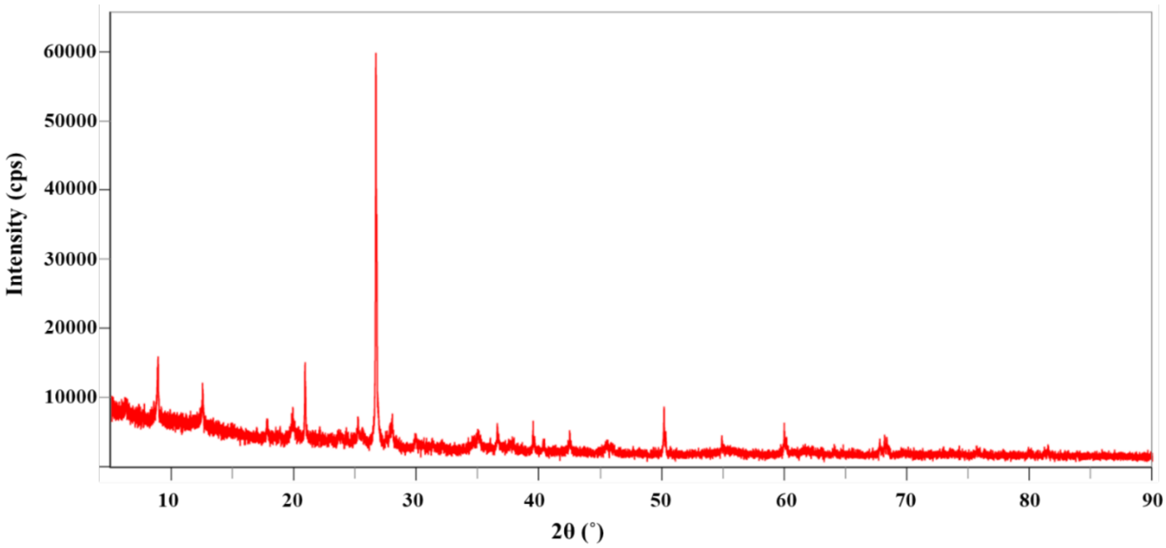


Fig. 4.4 : X-ray diffraction (XRD) pattern for the modified WTS.

Table 4.3. Mineralogical compositions in WTS and OPC found from XRD analysis

Type/ Parameters	Mineralogical Composition (%)			
	C_3S	C_2S	C_3A	C_4AF
Recommended values	50–65	10–25	9–12	7–11
WTS	23.43	40.35	9.08	17.14
Cement	54.72	10.26	3.56	13.53
Source	[117]			

The WTS exhibits significantly lower C_3S of 23.43% compared to the recommended range of 50-65%, and higher C_2S of 40.35% compared to the recommended range of 10-25%. The C_3A content in WTS is within the acceptable range, but C_4AF is notably higher than the recommended range. Additionally, the LSR for WTS is significantly lower (i.e., 0.011) than the recommended 0.80-0.96, while the SR and AR were within the recommended ranges as shown in **Table 4.4**. These discrepancies indicate that the WTS sample cannot be used alone as a cement in concrete and/or mortar production.

Table 4.4. Cement quality parameters in WTS and OPC.

Type/ Parameters	Cement Quality Parameters		
	LSR	SR	AR
Recommended values	0.80–0.96	1.7–2.7	0.90–1.9
WTS	0.011	1.220	1.022
Cement	0.86	2.33	3.12
Source	[27,121,122]		

Figure 4.5 illustrates a ternary diagram that maps the identified oxides available in different cementitious materials. It also demonstrates that WTS shares chemical characteristics similar to a natural pozzolans. Moreover, the CEM-I cement used in this study closely matches the chemical composition typically associated with Portland cement. This comparison suggests that both WTS and OPC/CEM-I have chemical profiles suitable for their intended uses.

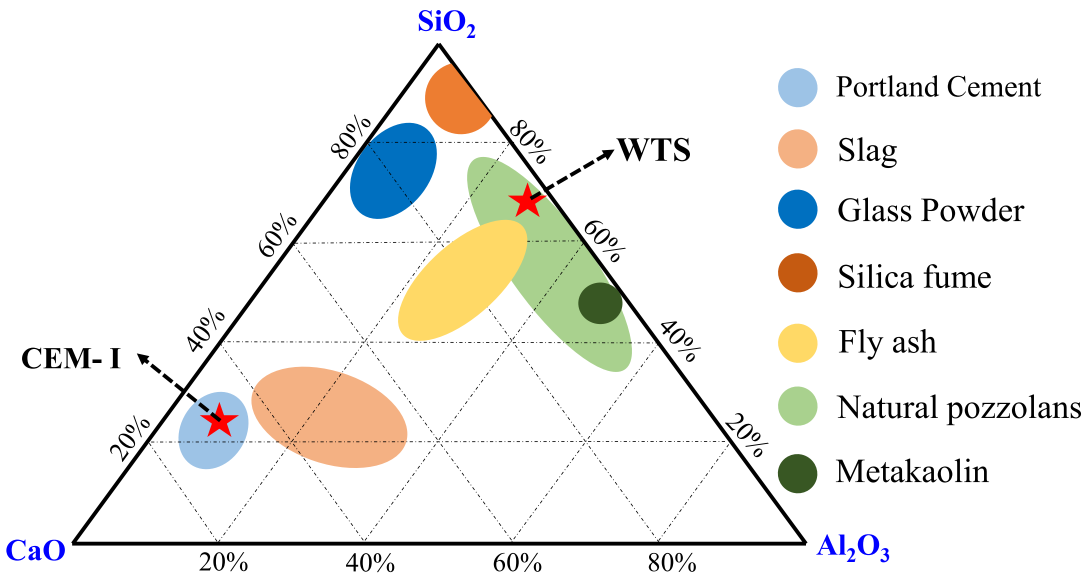


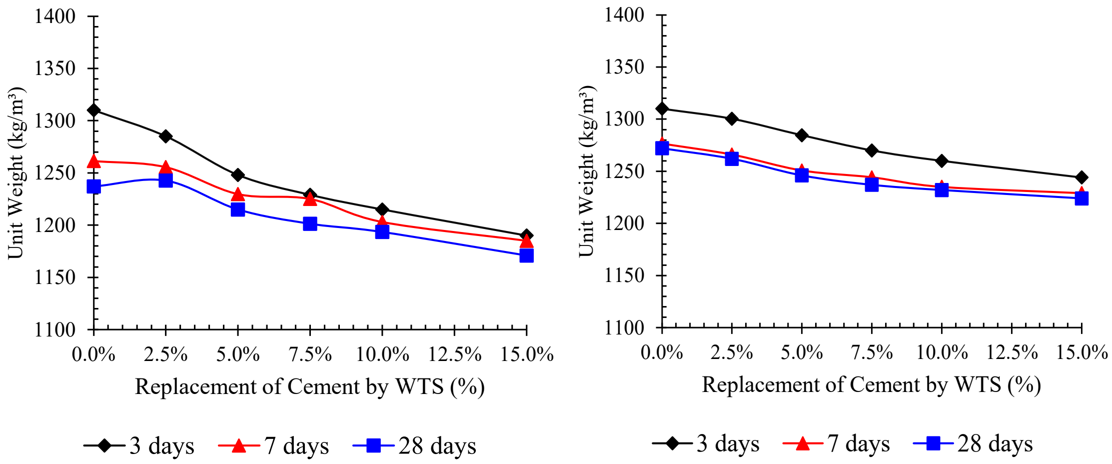
Fig. 4.5 : Ternary diagram for different cementitious materials [163–165].

Specifically, WTS exhibits traits akin to natural pozzolans, which are valued for their pozzolanic properties that enhance the durability and strength of concrete when blended with cementitious materials. On the other hand, CEM-I cement conforms to the standard composition expected of Portland cement, known for its widespread application in construction due to its reliable performance and versatility in various concrete applications.

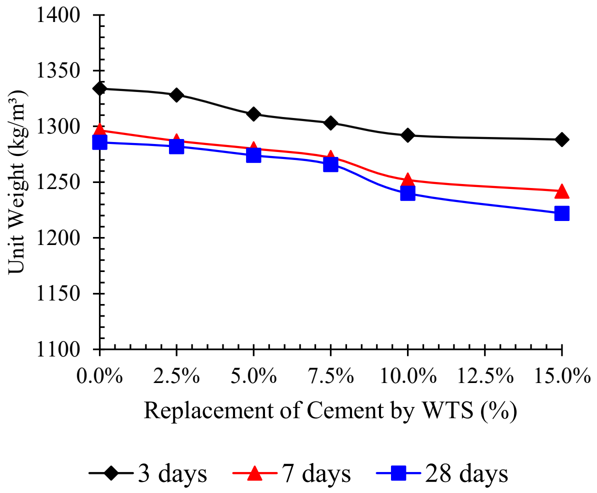
4.3 PERFORMANCE OF CEMENT MORTAR CONTAINING WTS

4.3.1 Variation of unit weight of cement mortar

Figure 4.6 shows the variations in unit weight of cement mortar when OPC was partially replaced by WTS for the mix ratios of 1:2, 1:4, and 1:6. The cement mortar with a 1:6 mix ratio consistently exhibits a higher unit weight, being 1.01 to 1.05 times heavier than the other mix ratios at 28 days of curing. This higher unit weight is due to the larger proportion of sand in the 1:6 ratio, followed by the 1:4 and 1:2 ratios, as the unit weight of sand is higher than the specific gravity of OPC and WTS [172,173]. The unit weights of cement mortar with a 1:2 mix ratio range from 1171 to 1310 kg/m³.



(a) (b)



(c)

Fig. 4.6 : Variation of unit weight for the cement mortar (a) 1:2, (b) 1:4 and (c) 1:6

While the unit weights of cement mortar with 1:4 and 1:6 mix ratios range from 1224 to 1310 kg/m³ and 1226 to 1334 kg/m³, respectively. These findings are lower than those reported by Dong et al. (2023), Nawab et al. (2023), and Shafiullah et al. (2015) where they replaced cement by slag, fly ash, rice husk ash, silica fume, metakaolin, coconut fibers, and lithium slag. Figure 4.6 shows that the unit weight of cement mortar consistently decreases with WTS addition level for all mix ratios. The unit weight dropped by about 3.44–4.95% for a 15% WTS addition level in a 1:6 mix, whereas the unit weight reduction was approximately 3.73–5.04% and 5.34–9.16% for 1:4 and 1:2 mix ratios, respectively. This decrease can be attributed to the lower unit weight of WTS compared to OPC cement, as well as the presence of a more porous C-S-H network and microcracks in the cement mortar, similar to cement concrete. The unit weight of the cement mortar was decreased significantly by 2.55–3.71% over the first 3–7 days. Afterward, the unit weight was continued to decrease but with lower rate ranging from 0.35 to 1.93% compared to the 7-day curing weights. A similar pattern of unit weight reduction was reported by Nawab et al. (2023), who replaced cement by silica fume, metakaolin, and coconut fibers in cement mortar [174]. These findings indicate that incorporating WTS can effectively reduce the overall weight of cement mortar, making it a promising option for lightweight cement paste applications.

4.3.2 Variation of compressive strength

Figure 4.7 illustrates the variation in compressive strength of cement mortar with WTS addition level, where cement mortars were prepared using three mix ratios of 1:2, 1:4, and 1:6. The cement mortar with a 1:2 mix ratio consistently demonstrates higher compressive strength, being 1.15–1.97 times stronger than the 1:4 ratio and 2.46–4.44 times stronger than the 1:6 ratio. The observed trend aligns with findings reported elsewhere [166–168]. The compressive strength at 28-days curing period of the 1:2 mix ratio mortar ranges from 31.66–40.22 MPa, while the 1:4 and 1:6 mix ratios exhibit ranges of 24.32–28.73 MPa and 11.54–15.34 MPa, respectively.

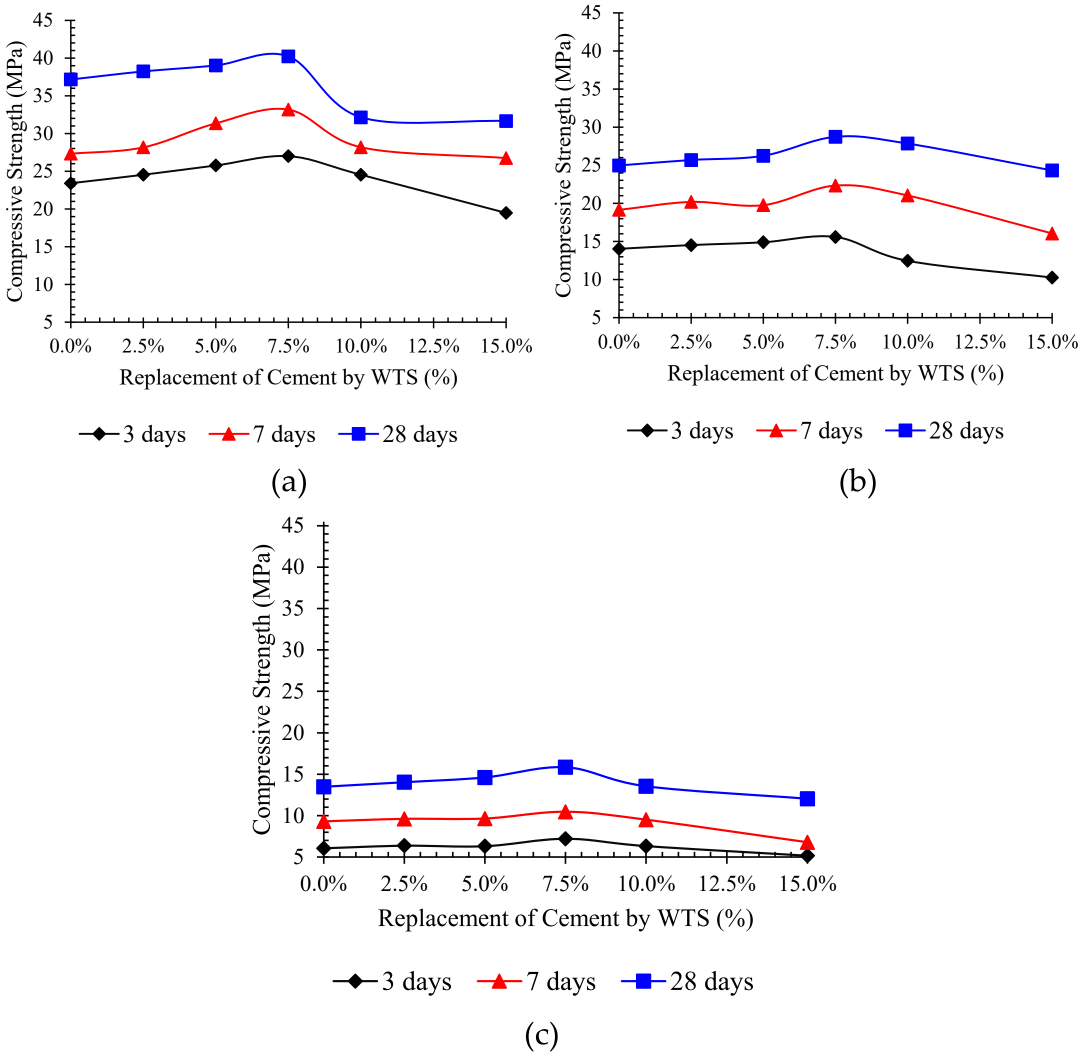


Fig. 4.7 : Variation of compressive strength for the cement mortar (a) 1:2, (b) 1:4 and (c) 1:6.

As depicted in **Figure 4.6**, the strength increases up to a 7.5% WTS addition level, followed by a steady decline beyond this point. The compressive strengths of cement mortars with 7.5% WTS were approximately 8%, 15%, and 18% higher than the control specimens for 1:2, 1:4, and 1:6 mix ratios, respectively. The optimal 7.5% WTS addition level in cement mortar is consistent with findings reported by Nega et al. (2023) [77], Woźniak et al. (2022) [69], El-Tair et al. (2018) [68], Chang et al. (2010) [80], and Naceri and Hamina (2009) [63], who used various waste powders (such as marble, granite, glass), sewage sludge ash, and waste brick as partial replacement of cement. The mechanism and factor for compressive strength development of cement mortars are similar to cement concrete. The higher compressive strength of the cement mortar with a 1:2 mix ratio compared to the 1:4 and 1:6 mix ratios can be attributed to several factors. The 1:2 mix ratio contains a higher proportion of cement compared to the 1:4 and 1:6 mix ratios. Cement is the primary binder in mortar, and a higher cement content generally leads to greater strength due to improved binding and filling of voids [169,170].

The water-cement ratio in the 1:2 mix is typically lower than in the 1:4 and 1:6 mixes. A lower water-cement ratio generally results in higher strength because it leads to better hydration of cement particles and reduced porosity in the hardened mortar [168,171]. The cement mortars reached 67.1%, 54.3%, and 43.7% of their compressive strength within the first 3 days for the 1:2, 1:4, and 1:6 mix ratios, respectively, at a 7.5% WTS replacement. In addition, the cement mortars attained 82.5%, 77.8%, and 65.0% of their ultimate strength by 7 days. These results suggest that cement mortars undergo significant early strength development within the first 7 days, with the 1:2 mix outperforming the 1:4 and 1:6 mix ratios during this initial period.

4.3.3 Variation of flexural strength

Figure 4.8 depicts the changes in flexural strength of cement mortar with WTS addition level after 28 days curing period. The values of flexural strength were ranged from 4.64 MPa to 5.88 MPa for the studied WTS addition range. Initially, the flexural strength was increased almost linearly up to the WTS addition level of 5% with an increasing rate of 12.22% approximately. The flexural strength began to decrease beyond the 5% addition level, dropping to 4.64 MPa at a 15% WTS level. The flexural strength results are consistent with other findings where waste granite, glass powder, textile sludge, and sewage sludge ash were used as partial substitutes [11,63,67–69].

The variation in flexural strength with different proportions of WTS replacement can be attributed to several factors. At lower replacement levels (0% to 5%), WTS particles may effectively fill voids and enhance the strength of the cement mortar. However, at higher replacement levels (beyond 5%), the larger or irregularly shaped WTS particles can disrupt the cement matrix and affect hydration reactions negatively [146,175].

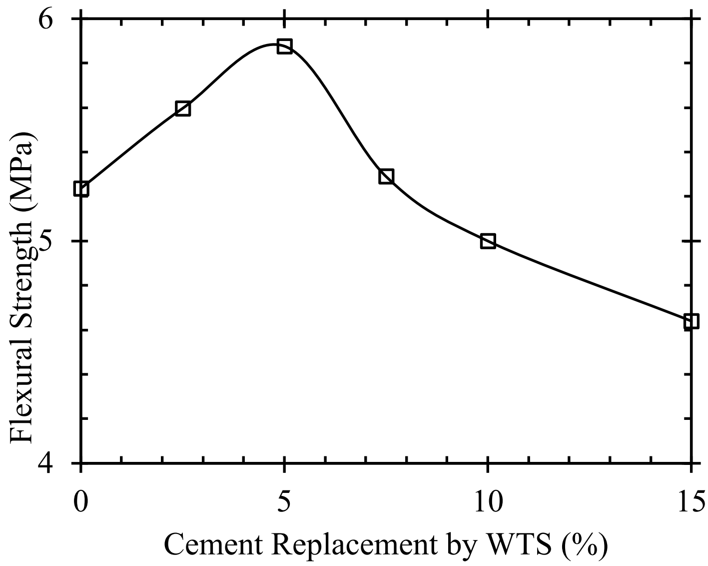


Fig. 4.8 : Flexural strength variation of cement mortar with mix ratio of 1:4 at 28 days of curing.

The chemical composition of WTS might also interfere with cement hydration processes, while excessive replacement could dilute the pozzolanic effect and alter the water-cement ratio, reducing strength [176–178]. Additionally, WTS might initially improve bonding and workability, but excessive amounts could compromise these properties and the overall microstructural integrity of the mortar.

4.4 PERFORMANCE OF CONCRETE WITH WTS REPLACEMENT

4.4.1 Effects on workability of concrete

The slump test was performed for each concrete mixture to assess the workability of concrete mixture. **Figure 4.9** depicts the changes in slump values when WTS is partially substituted for cement in various proportions for concrete mix ratios of 1:2:4 and 1:1.5:3. The slump test is a measure of the workability of concrete, with higher values indicating a more fluid mix that is easier to handle, place, and compact. The range of slump values for the concrete mixes varies from 110 mm to 145 mm. The slump values range from 120 mm to 140 mm for the 1:2:4 concrete mix without any admixture, while the same mix with admixture has values ranging from 130 mm to 145 mm. Similarly, the 1:1.5:3 concrete mix without admixture shows slump values between 110 mm and 135 mm, and the range is from 120 mm to 140 mm with admixture. The slump values found in this study align with those reported by different researchers [58,60], who investigated the use of lime sludge and waste paper sludge as partial cement replacements in concrete. In contrast, the slump values observed here are higher in this study than those reported elsewhere, where sewage sludge and waste paper pulp were used as partial replacements for cement [71,75].

Figure 4.9 also demonstrates that the slump values rise with the incorporation of WTS as a partial replacement for cement in concrete. The slump values for concrete mixes with a 1:2:4 ratio increase by 15-20 mm when WTS is added in proportions ranging from 0–15%. Moreover, concrete mixes with a 1:1.5:3 ratio show a more significant increase in slump values, ranging from 20 to 25 mm. The results indicate that adding WTS to concrete as a partial replacement for cement improves the workability of the concrete, as reflected by the higher slump values in both mixes. The continuous increase in slump value with the addition of WTS as a partial replacement of cement in concrete can be attributed to several factors. WTS contains fine particles that act as fillers, improving the overall workability of the concrete mix by occupying interstitial spaces between larger aggregates [179].

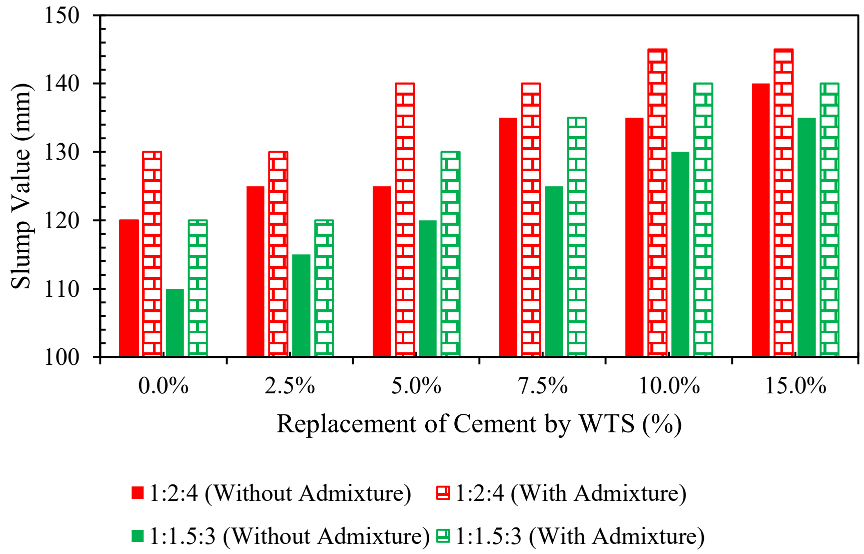


Fig. 4.9 : Variation of slump values of concrete with replacement of cement by WTS.

Moreover, the excessive addition of WTS might alter the hydration process of cement, which could potentially facilitate the formation of a more porous C-S-H network. This enhanced porous network improved the flow of water within the concrete, thereby increasing its slump value [180–182]. Furthermore, the microcracks that developed due to the addition of WTS to the concrete might contribute further to the observed increase in slump value. The 1:2:4 mix consistently demonstrates higher slump values than the 1:1.5:3 mix. For instance, the slump value for the 1:2:4 mix is 120 mm at 0.0% admixture, while it is 110 mm for the 1:1.5:3 mix. The slump values for the 1:2:4 concrete mix ratio are 5-10 mm higher than those for the 1:1.5:3 concrete mix ratio. This trend continues at higher percentages of admixture, with the 1:2:4 mix showing higher workability. The use of admixtures has a notable impact on the workability of concrete.

As seen **in Figure 4.9**, the inclusion of admixtures increases the slump value across all mix ratios. For instance, at a 10.0% admixture level, the slump value for the 1:2:4 mix increases from 135 mm (without admixture) to 145 mm (with admixture). Similarly, for the 1:1.5:3 mix, the slump value increases from 130 mm (without admixture) to 140 mm (with admixture). The difference in slump values is approximately 5-15 mm for the 1:2:4 concrete mix ratio, whereas for the 1:1.5:3 concrete mix ratio, the difference is around 5-10 mm. Admixtures impact concrete workability by adjusting water content, air entrainment, setting time, and rheological properties, influencing slump values accordingly [183–185]. The results demonstrates that admixtures make the concrete mix more workable, which can be particularly beneficial in applications requiring easy placement and compaction.

4.4.2 Effects on unit weight of concrete

The unit weight of cubical concrete specimen was measured at different concrete age such as 7, 14, 28, and 72 days. **Figure 4.12** illustrates the variation in unit weight when WTS is partially replaced with cement in different proportions over various curing periods for concrete cube mix ratios of 1:2:4 and 1:1.5:3. The 1:1.5:3 concrete mix consistently demonstrates higher unit weight, being 1.03 to 1.10 times heavier than the 1:2:4 mix. The unit weights range from 2160 to 2489 kg/m^3 for the 1:2:4 concrete mix ratio, while the 1:1.5:3 concrete mix ratio ranges from 2350 to 2567 kg/m^3. The unit weight of concrete is consistent with the findings of Krishta David & Karan Nair (2018) and Opara et al. (2018) [51,197], who replaced sewerage sludge and dust in concrete at different curing durations and proportions, which ranged from 2382 to 2508 kg/m^3. Moreover, since the specific gravity of WTS is approximately 27.5% lower than OPC cement, incorporating additional WTS as a partial replacement may lead to a decrease in unit weight [198].

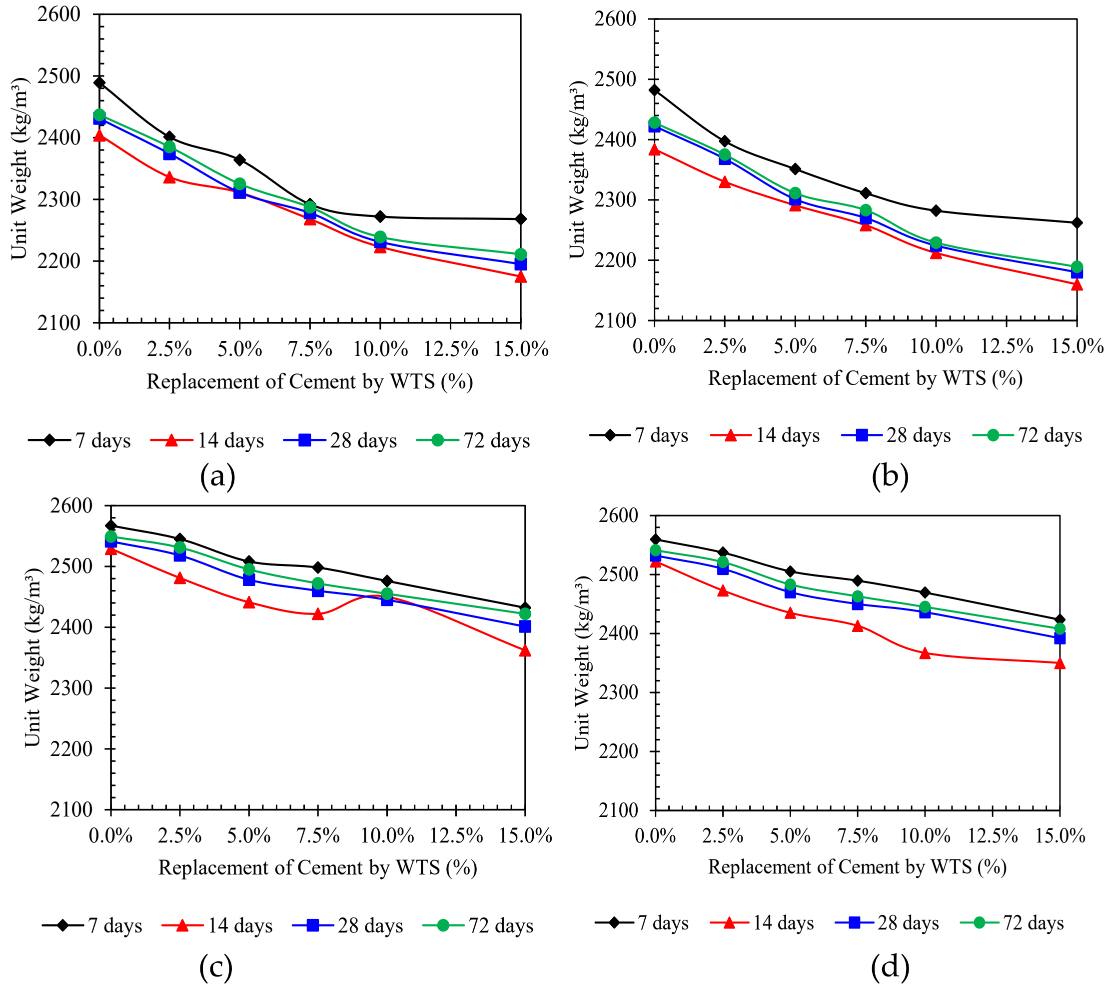


Fig. 4.10 : Variation of Unit weight of cube specimen (a) 1:2:4 (without admixture), (b) 1:2:4 (with admixture), (c) 1:1.5:3 (without admixture), (d) 1:1.5:3 (with admixture)

In addition, the addition of WTS in concrete can result in a porous C-S-H network and the development of micro cracks, which may also contribute to a reduction in unit weight as discussed earlier. Furthermore, the incorporation of an admixture resulted in a reduction of unit weight by approximately 4-22 kg/m^3 for the 1:2:4 concrete mix, while the 1:1.5:3 concrete mix experienced a more pronounced decrease, ranging from 3-83 kg/m^3. As depicted in **Figure 4.12**, the unit weight consistently decreased with the increasing amount of WTS used as a partial replacement for cement in the concrete. The results also indicate that both mixes experience a significant decrease in unit weight within the first 14 days, with the 1:2:4 mix showing slightly lower unit weight than the 1:1.5:3 mix during this initial period. The unit weight decreased by approximately 9.5% at higher replacement levels for the 1:2:4 mix, whereas the 1:1.5:3 concrete mix showed a reduction of around 5.7%. Initially, the unit weight of the 1:2:4 concrete mix decreased by 1.0-4.5% within the first 14 days, and the 1:1.5:3 mix decreased by 1.1-4.1%. Subsequently, the unit weight of both mixes increases slightly for further curing days, with changes ranging from 0.2 to 2.1% compared to the unit weights at 7 and 72 days of curing. A similar trend in unit weight changes was reported by Krishta David & Karan Nair (2018) [51], where sewerage sludge was partially substituted for cement in concrete. These findings suggest that the incorporation of WTS can effectively reduce the overall weight of concrete, making it a viable option for lightweight concrete applications.

Figure 4.13 illustrates the variation in unit weight observed after a 28 days curing period for concrete cylinders, with different proportions of WTS replacing cement in mix ratios of 1:2:4 and 1:1.5:3. Both concrete mixes showed a steady decrease in unit weight with higher WTS content. The unit weight of cylindrical specimens was 4.94% higher than that of cubic specimens.

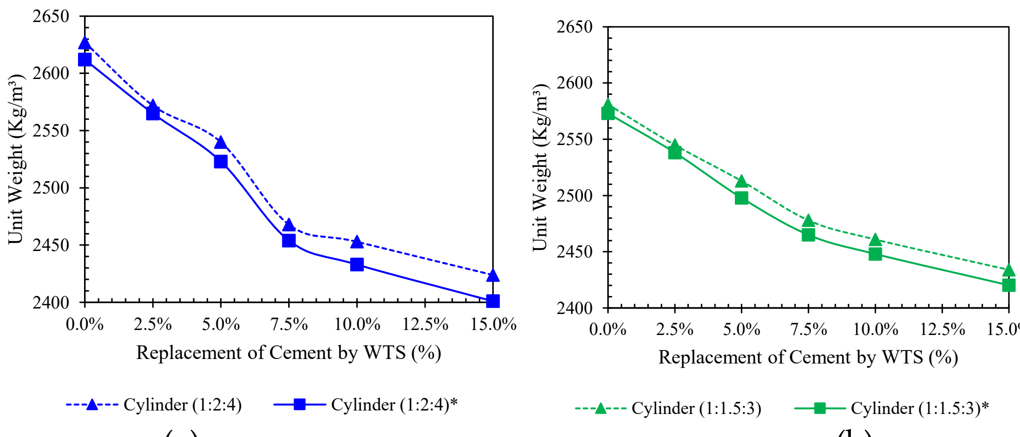


Fig. 4.11 : Variation of Unit weight of cylinder (a) 1:2:4, (b) 1:1.5:3 (* mark indicates concrete cylinder made without any addition of admixture)

The unit weights of cylindrical specimens for both mixes were almost similar, with the 1:2:4 concrete mix ranging from 2401 to 2627 kg/m^3 and the

1:1.5:3 concrete mix ranging from 2420 to 2581 kg/m^3. The addition of an admixture resulted in an increase in unit weight for both mix ratios. The increase was 7-20 kg/m^3 for the 1:2:4 concrete mix, while the 1:1.5:3 mix showed a more substantial increase of 7-20 kg/m^3.

4.4.3 Variation of compressive strength of concrete

To investigate the influence of WTS on the compressive of concrete, the concrete 100 mm cubical specimens were made considering two mix ratios of 1:2:4 and 1:1.5:3 with a fixed water cement ratio of 0.45. To attain the desired workability of the concrete mixture, Superplasticizer admixture was added in the mixture. The compressive strengths were determined at different curing ages of the concrete. **Figure 4.12** shows the variation of strength when WTS is partially replaced with cement in different proportions over various curing periods for concrete cube mix ratios of 1:2:4 and 1:1.5:3. The concrete with a 1:1.5:3 mix ratio consistently exhibits higher compressive strength, being 1.03–1.45 times stronger than the 1:2:4 ratio. The 1:2:4 concrete mix ratio shows compressive strength values ranging from 11.67–21.04 MPa, while the 1:1.5:3 concrete mix shows a range of 10.6 to 21.5 MPa. The compressive strength of the 1:2:4 concrete mix increased by 3.6–19.6% with the addition of an admixture, reaching 13.2–24.10 MPa.

In contrast, the 1:1.5:3 concrete mix saw a more significant increase of 26.6% to 64.3%, achieving a compressive strength of 15.27–33.41 MPa. The results of compressive strength are consistent with those reported by different literature [29,54,71–73,75], where the reported compressive strengths ranged between 20–40 MPa when using sewage sludge, textile sludge, tannery sludge, and waste paper pulp as partial replacements for cement in concrete.

As seen in **Figure 4.12,** the strength increased up to a 5% WTS replacement level with cement, followed by a continuous decline beyond this point. The strength decreased by approximately 26% at higher replacement levels for the concrete, with a mix ratio of 1:2:4. In contrast, the concrete with a mix ratio of 1:1.5:3 showed a reduction in strength of around 37%.

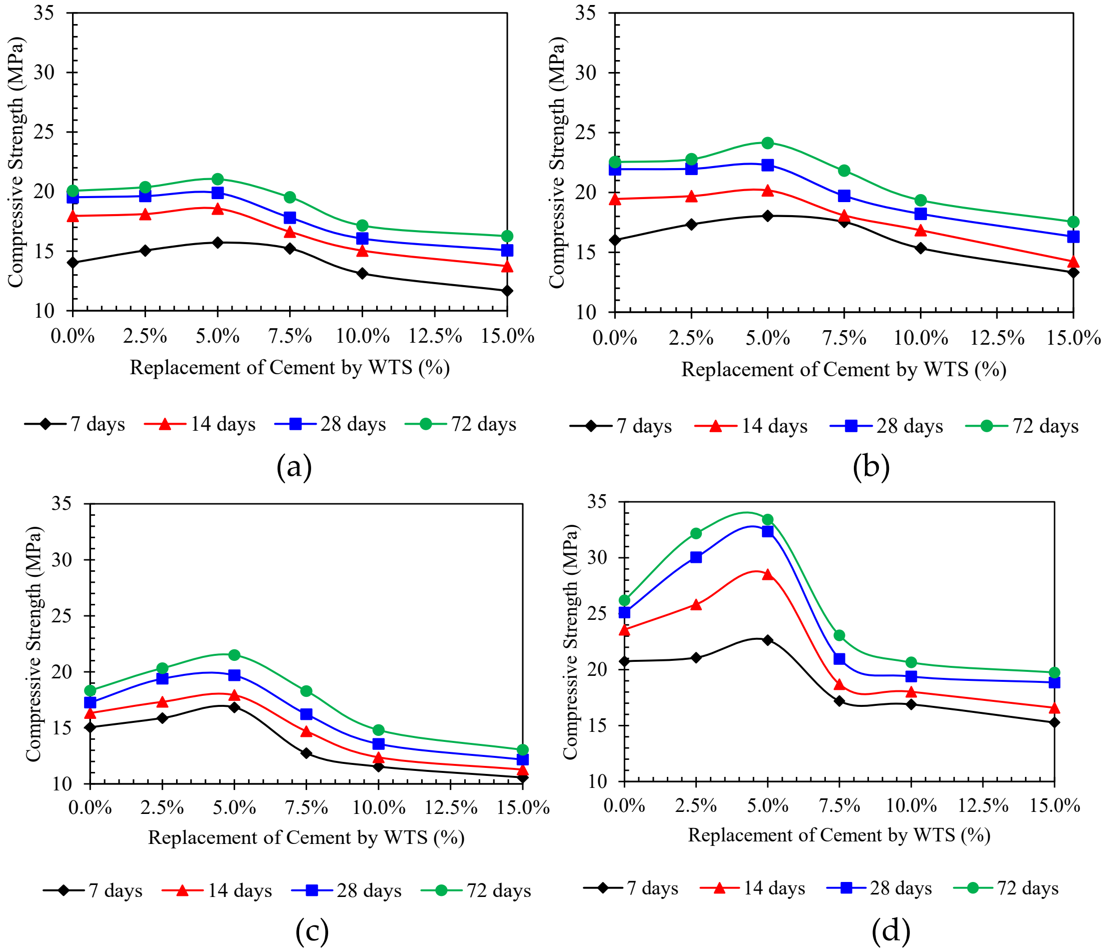


Fig. 4.12 : Variation of Concrete Compressive Strength (a) 1:2:4 (without admixture), (b) 1:2:4 (with admixture), (c) 1:1.5:3 (without admixture), (d) 1:1.5:3 (with admixture).

The optimal amount of 5% WTS as a partial cement replacement in concrete aligns with the results reported by Goyal et al. (2019) and Teja et al. (2017) [11,57] for using textile sludge and hypo sludge as partial replacement of cement. The 1:2:4 concrete mix ratio achieved 69.7% to 80.4% of its compressive strength within the first 7 days. In comparison, the 1:1.5:3 concrete mix ratio showed a compressive strength gain of 65.5% to 82.1%. On the other hand, the 1:2:4 concrete mix had attained 90.4% to 97.4% of its ultimate strength by 28 days, whereas the 1:1.5:3 concrete mix had reached 88% to 96.8% of its final compressive strength. The results indicate that both concrete mix ratios experience substantial early strength development within the first 7 days, with the 1:2:4 mix slightly outperforming the 1:1.5:3 mix in this initial period. However, both mixes achieve nearly their full-strength potential by the end of 28 days, with the 1:2:4 concrete mix again showing a marginally higher percentage of strength gain compared to the 1:1.5:3 mix. The optimal replacement proportion of WTS with cement in concrete was determined to be 5%, at which point the compressive strength was approximately 12% higher for the 1:2:4 concrete mix and 10% higher for the 1:1.5:3 concrete mix compared to control specimens.

Figure 4.13 shows the variation in compressive strength observed after a 28-day curing period for concrete cylinders, with different proportions of WTS replacement with cement having mix ratios of 1:2:4 and 1:1.5:3. The compressive strength of cubic specimens was consistently found to be approximately 1.14 times higher than that of cylindrical specimens. This trend mirrored the observations made for cubic specimens, indicating a similar behavior across both types of concrete test samples. Concrete mixed with a ratio of 1:1.5:3 consistently demonstrated higher compressive strength compared to the 1:2:4 concrete ratio, ranging from 1.01–1.05 times stronger. The 1:2:4 mix exhibited compressive strength values ranging from 11.95–18.09 MPa, while the 1:1.5:3 mix ranged from 12.24–18.40 MPa.

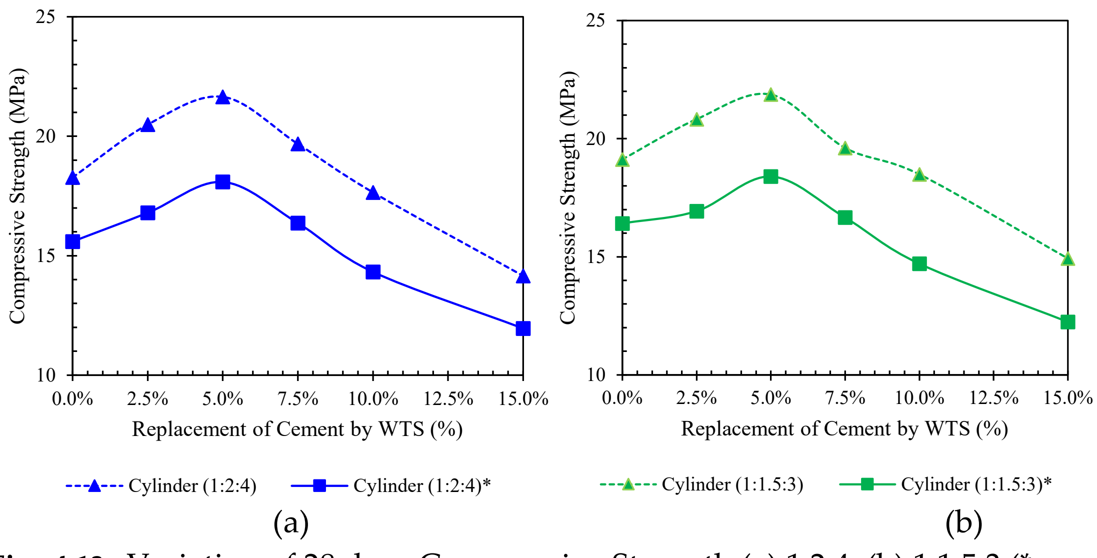


Fig. 4.13 : Variation of 28 days Compressive Strength (a) 1:2:4, (b) 1:1.5:3 (* mark indicate the casting of concrete was made without addition of any admixture)

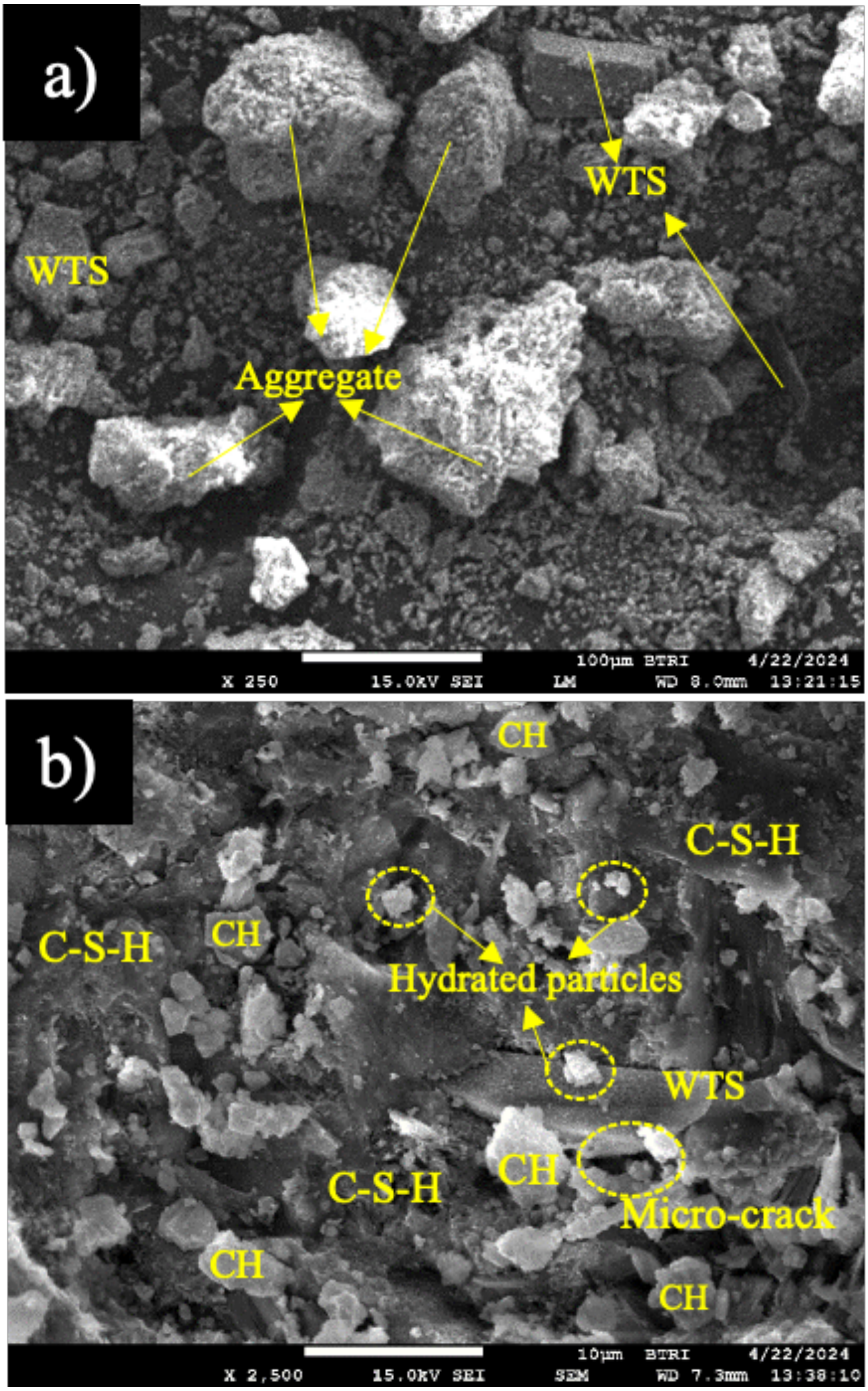
The addition of an admixture resulted in a significant increase in compressive strength for both mix ratios. The 1:2:4 mix ratio had an increase of 14.69% to 18.86%, achieving compressive strengths ranging from 14.15–21.65 MPa. In contrast, the 1:1.5:3 mix showed a more substantial improvement of 14.17% to 20.45%, reaching compressive strengths of 14.92–21.87 MPa. As seen in **Figure 4.13**, compressive strength increased up to a 5% WTS replacement level for both concrete mix ratiso before declining continuously thereafter.

The optimal 5% replacement leverages the high silica content for pozzolanic activity, filling voids, and enhancing particle packing density. Again, WTS is characterized by its high silica content (see **Table 4.2** and **4.4**), particularly in forms of C_2S and C_3S, which exhibit pozzolanic properties suitable for concrete applications. When WTS is incorporated as a partial substitute for cement, WTS reacts with $Ca(OH)_2$ produced during hydration to form additional C-S-H gel. This gel fills voids between aggregate particles, enhancing packing density and bonding within the concrete, thereby improving both early and long-term compressive strength and durability [186–188]. WTS also contains significant amounts of Al_2O_3 (18.80%) and Fe_2O_3 (18.40%), which influence the hydration process and contribute to the overall performance of the concrete [189–191].

Several factors contribute to decline in compressive strength when WTS addition level is more than 5%. Higher WTS addition levels introduce an excessive amount of fine particles, disrupting workability and compaction, and thereby resulting in incomplete reactions and weaker concrete. The excess silica content in WTS addition level beyond optimum WTS addition level (i.e., 5%) can react much during hydration, potentially leading to excessive CH along with C-S-H gel [192,193]. Although CH helps maintain the necessary alkalinity for concrete durability, an excess can pose durability challenges and reduce strength [187,188,193]. Moreover, initial formation of C-S-H begins with nucleation on the surfaces of cement particle, followed by C-S-H fibril growth that creates a porous network [192,194]. However, some porosity is needed for hydration and strength development, but excessive porosity weakens the concrete. In addition, the low calcium oxide (CaO) content in WTS (1.74%) limits the formation of essential hydration products, further reducing overall strength. Increased amount of C_4AF and alkaline content (Na_2O and K_2O) in WTS can cause micro-cracks due to stress concentrations, which also degrade the compressive strength of concrete [192,195,196]. Although the Loss on Ignition (LOI) value of WTS is low (0.17%), higher WTS content can lead to lower compressive strength due to the potential decomposition of organic matter during mixing or hydration, further compromising structural integrity [11].

4.4.4 Microstructural changes of concrete due to WTS replacement

From the above discussions, it is observed that 5% WTS addition level provides maximum compressive strength, good workability and reasonable unit weight of concrete. Therefore, a SEM analysis was conducted on a concrete sample containing 5% WTS addition level. **Figure 4.14** presents the microscopic images of the concrete sample. **Figure 4.14(a), (b), (c)**, and **(d)** represent the images at 250, 2500, 10000, and 25000-times magnifications, respectively. Aggregates and WTS can be observed at lower magnification. Various hydration products, such as Calcium Silicate Hydrate (C-S-H), Calcium Hydroxide (CH), and small hydrated particles, are also identified in the SEM images of concrete. In addition, several micro-cracks are also visible. The alkaline content (Na_2O and K_2O) in both OPC and WTS (see **Table 4.2**) reacts with reactive silica in the aggregates, forming a gel that absorbs water and expands. This expansion can generate internal stresses, potentially causing micro-cracks in the concrete [195,199].


a)
WTS
WTS
Aggregate
100µm BTRI 4/22/2024
X 250 15.0kV SEI LM WD 8.0mm 13:21:15
b)
CH
C-S-H
C-S-H
CH
Hydrated particles
WTS
CH
C-S-H
Micro-crack
CH
CH
10µm BTRI 4/22/2024
X 2,500 15.0kV SEI SEM WD 7.3mm 13:38:10

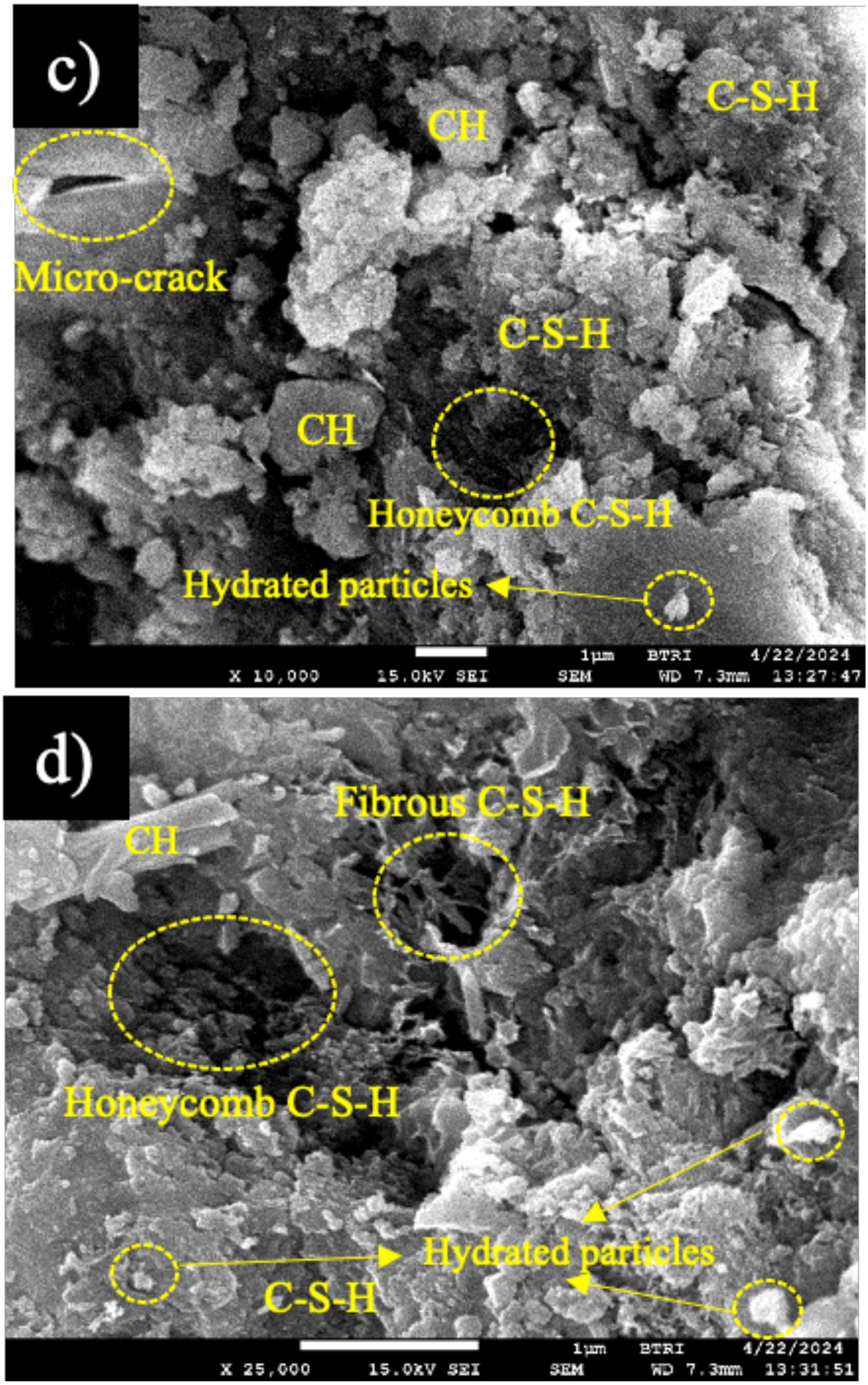


Fig. 4.14 : SEM morphologies of concrete partially replaced by cement with WTS at (a) 250, (b) 2,500, (c) 10,000, (d) 25, 000 times magnification.

The formation of C-S-H and CH during the chemical reactions primarily involve C_2S and C_3S [186,200,201], in OPC and WTS as outlined in **Table 4.3**. A series of chemical reactions, known as hydration, occur when water is added during concrete casting. The main reactions involve the silicate phases (C_2S and C_3S) in OPC and WTS with water as shown follows:

$$2C_3S+6H_2O \rightarrow 3CaO \cdot 2SiO_2 \cdot 3H_2O\ (C-S-H) + 3Ca(OH)_2\ (CH)$$

$$2C_2S+4H_2O \rightarrow 3CaO \cdot 2SiO_2 \cdot 3H_2O\ (C-S-H) + Ca(OH)_2\ (CH)$$

The C-S-H contributes to the dense microstructure of concrete, reducing porosity and enhancing durability (**Figure 4.14(b)** and **(d)).** CH, also known as portlandite, is formed as a byproduct of the hydration reactions in the silicate phases (C_3S and C_2S) [187,202]. CH maintains alkalinity (pH) but can also pose durability challenges and strength reduction if it is present in excess amount [187,188,193].

The initial formation of C-S-H begins with nucleation on the surface of cement grains, followed by the growth of C-S-H fibrils (see **Figure 4.14(d)**). These fibrils intertwine to create a porous network, which can appear honeycomb-like under a microscope, as shown in **Figures 4.14(c)** and **(d)**. The spaces between the C-S-H fibrils contribute to the overall porosity of the concrete and become more defined as hydration progresses, enhancing the honeycomb appearance [194,203]. Additionally, the admixture used in this study disperses cement particles more effectively, promoting more uniform hydration and potentially more distinct honeycomb structures [196]. Moreover, WTS, containing C_2S and C_3S, acts as a pozzolanic material and reacts with CH to form additional C-S-H [204], further altering the microstructure and enhancing the honeycomb appearance. The spaces within the honeycomb structure and fibrous C-S-H contribute to the overall porosity of the concrete. Although some porosity is necessary for hydration and strength development, but excessive porosity can weaken the concrete [205,206].

4.5 CORRELATION AND PRINCIPAL COMPONENT ANALYSIS (PCA)

Figure 4.15 shows a correlation plot that highlights the relationships among various concrete variables. It reveals a significant positive correlation among the proportion of WTS replacement and both the water-cement ratio and slump value, indicating that both the water-cement ratio and slump increase as WTS is added, as also shown in **Figure 4.9**. This correlation suggests that the workability of the concrete improves continuously with the inclusion of WTS as a partial cement replacement, consistent with previous discussions.

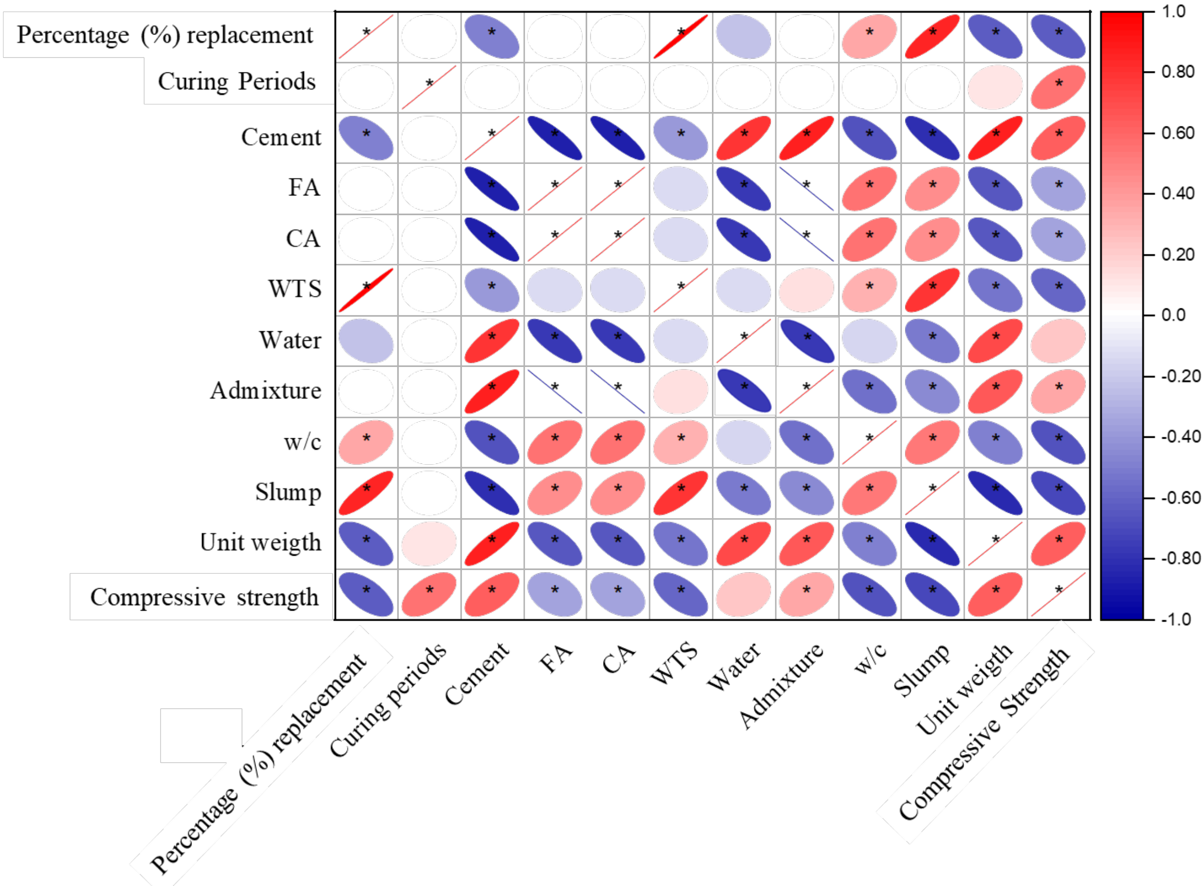


Fig. 4.15 : Correlation plot of different variables for the concrete specimens.

In contrast, there is a notable negative correlation between the proportion of WTS and both unit weight and compressive strength. This indicates that, overall, increasing the amount of WTS as a cement replacement tends to decrease the compressive strength of the concrete. However, it is important to note that at certain proportions, WTS lead to an increase in compressive strength (see **Figure**

4.12), suggesting that the effect is not uniform across all levels of replacement. Additionally, the negative correlation with unit weight implies that incorporating WTS into the cement mix significantly reduces the concrete's weight. This reduction in weight highlights the potential for using WTS to produce lightweight concrete, offering benefits in applications where lower weight is advantageous. **Figure 4.15** also illustrates the effects of using admixtures in concrete. The addition of an admixture shows a positive correlation with compressive strength, unit weight, and cement proportion, suggesting that it enhances these properties. On the other hand, there is a significant negative correlation with water content and water-cement ratio, indicating that the admixture reduces both the water content and the water-cement ratio. This reduction, combined with its role as a water-reducing agent and superplasticizer, contributes to an increase in compressive strength in the concrete mix.

Figure 4.16 illustrates the correlations among various cement mortar variables, including the constituent materials, strength, curing periods, and the proportion of WTS used as a replacement for cement. The plot shows a significant negative correlation between the proportion of WTS and the unit weight of the mortar, indicating that as the WTS content increases, the unit weight of the mortar decreases substantially. This reduction in unit weight is a direct effect of incorporating WTS.

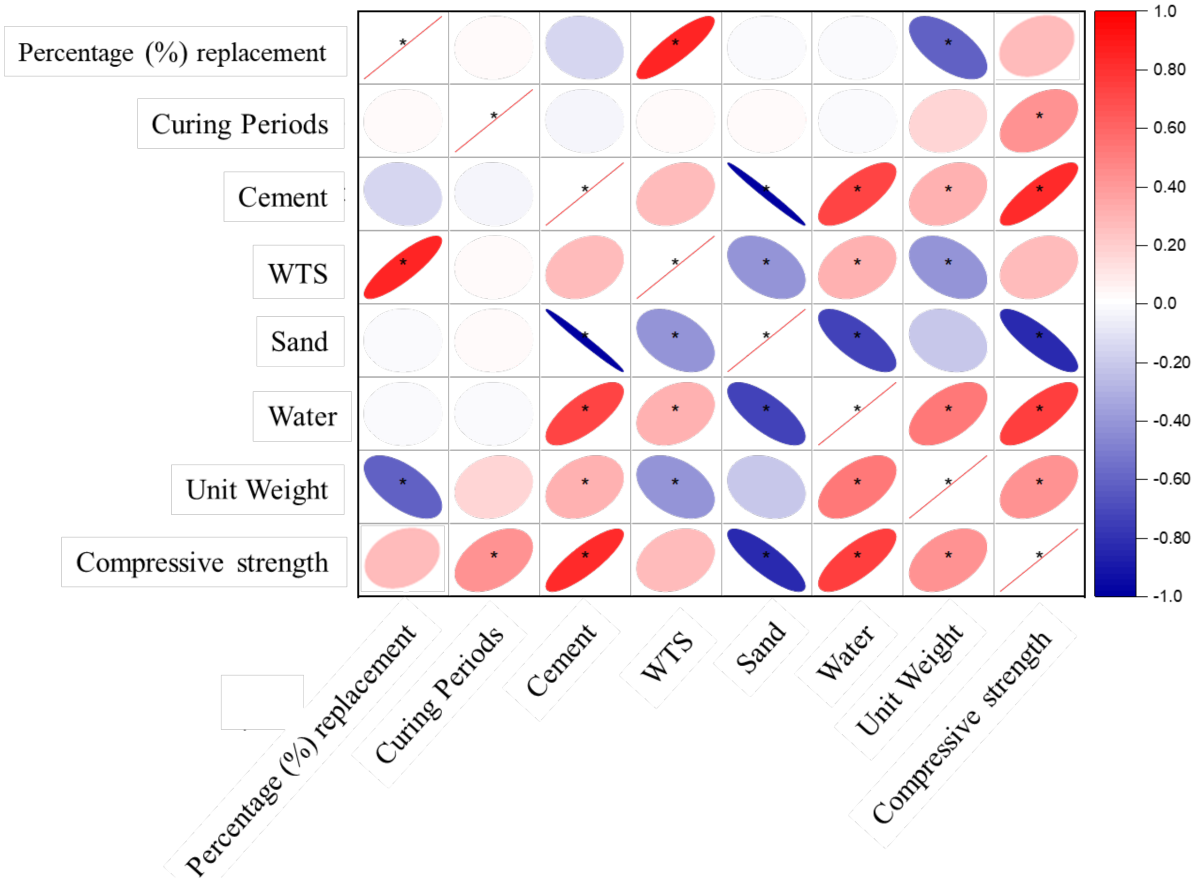


Fig. 4.16 : Correlation plot of different variables for the cement mortar specimens.

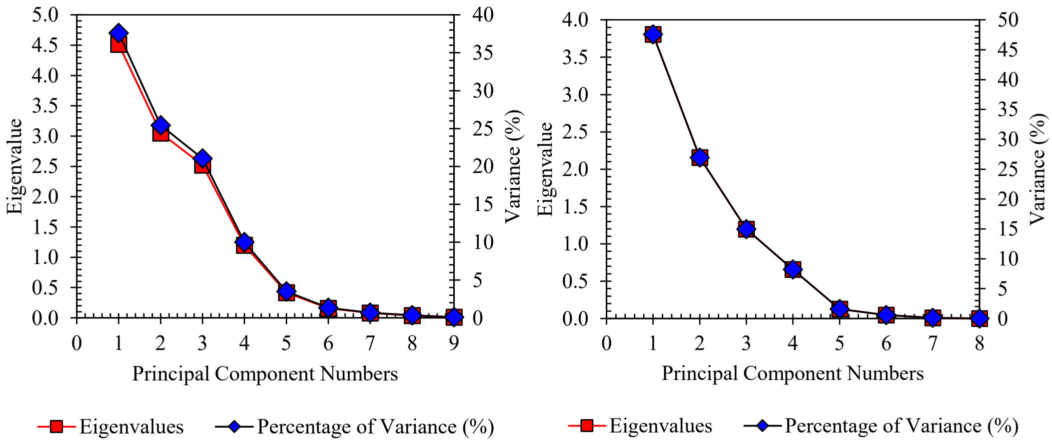
On the other hand, the proportion of WTS exhibits a positive relationship with compressive strength, suggesting that the compressive strength of the mortar tends to increase with higher WTS content up to certain proportion. However, this relationship is not statistically significant, meaning that while there is a general trend of increasing compressive strength with WTS addition, the magnitude of this increase is relatively small. The study also observed that a 7.5% replacement of cement with WTS enhances compressive strength, but further increases in WTS proportion result in a decline in compressive strength. These findings suggest that WTS can be effectively used to create more sustainable and lighter cement mortar, benefiting applications where reduced weight is advantageous.

The data sets' suitability for Principal Component Analysis (PCA) was evaluated using the Kaiser-Maier-Olkin (KMO) measure and Bartlett's sphericity test as shown in **Table 4.5**. For PCA to be appropriate, the KMO value should exceed 0.5, and the significance value of Bartlett's test should be below 0.05 [207,208]. The cement mortar dataset showed a KMO value of 0.672, while the concrete dataset had a KMO value of 0.512, indicating marginal sampling adequacy for PCA.

Table 4.5. Results of Kaiser-Maier-Olkin (KMO) Measure and Bartlett's Sphericity Test for Assessing PCA Suitability.

Statistical Variables		Cement Mortar	Concrete
Kaiser-Meyer-Olkin Measure of Sampling Adequacy		0.672	0.512
Bartlett's Test of Sphericity	Approx. Chi-Square	3.729	10.267
	df	1	1
	Sig.	0.05	0.001

Figure 4.17 illustrates the eigenvalues and percentage of variance for each principal component, highlighting its significance in capturing the variability of the dataset. Principal Component 1 (PC1) has eigenvalues of 4.51 and 3.81 with 37.62% and 47.59% of the total variance for cement cement mortar and concrete variables, respectively, making it the most influential component. Principal Component 2 (PC2) is also significant based on the variables, as it shows an additional 25.41% and 26.96% of the variance with eigenvalues of 3.1 and 2.2 for cement mortar and concrete, respectively, while Principal Component 3 (PC3) accounts for 21.03% and 14.97% of the variance, with eigenvalues of 2.52 and 1.19. In total, PC1, PC2, and PC3 together make up approximately 84.06% and 90.10% of the total variance for cement mortar and concrete, respectively. Principal Component 4 contributes an additional 10.00% and 8.20% of the variance, with subsequent components explaining less than 10.59% and 9.90% of the variance. Thus, retaining the first three principal components is adequate for simplifying the data while maintaining its most significant patterns.



(a) Cement Concrete (b) Cement Mortar

Fig. 4.17 : Scree plot displaying the distribution of principal components by their eigenvalues and percentage of variance for the variables

PCs is crucial for understanding how the addition of various replacement materials and WTS affects the performance characteristics of cement mortar and concrete. **Table 4.6** presents the factor loadings for principal components derived from variables related to both cement mortar and concrete. PC1 in the PCA is notably influenced by the proportion of replacement materials and WTS, highlighting its role in detaining how these additives affect the cement mortar and concrete mix.

Table 4.6. The factor loadings for the principal components in PCA that were derived from the variables of both cement mortar and concrete

Parameters	**Concrete**			**Cement Mortar**		
	PC1	**PC2**	**PC3**	**PC1**	**PC2**	**PC3**
Proportion (%) of WTS addition	**0.483**	-0.125	0.286	**0.628**	0.047	0.224
Curing periods	0.074	0.025	-0.125	-0.134	0.099	**0.844**
Cement	0.107	**0.453**	-0.045	-0.034	**0.474**	-0.247
FA	-0.160	0.233	**0.504**	-0.019	**-0.480**	0.228
CA	-0.160	0.233	**0.504**	-	-	-
WTS	**0.501**	-0.044	0.282	**0.568**	0.245	0.116
Water	-0.098	**0.423**	0.186	-0.107	**0.443**	-0.110
Admixture	0.176	**0.440**	-0.004	-	-	-
Water-cement ratio	**-0.511**	-0.170	0.086	-	-	-
Slump	0.077	**-0.309**	**0.429**	-	-	-
Unit weight	-0.192	**0.343**	-0.084	**-0.502**	0.190	0.212
Compressive strength	**0.322**	0.230	-0.281	0.313	**0.490**	0.231

The significant positive loadings for these variables suggest that PC1 reflects the variability introduced by incorporating different replacement materials and WTS into the mix. Specifically, PC1 reflects how the use of these materials impacts various properties of the mix, such as its workability and overall composition. The negative correlation of PC1 with unit weight indicates that as the proportion of replacement materials and WTS increases, there is a corresponding decrease in density (unit weight) of the concrete or cement mortar. This suggests that while these additives might influence the mix in terms of workability and other properties, they tend to reduce the overall weight. This reduction could be due to the lower density of the replacement materials or changes in the material's microstructure caused by the additives, which impact the binding and strength properties of the mix. PC2 predominantly reflects the core properties and mix components, showing low to moderate positive loadings for cement, water, admixture, and unit weight, and negative loadings for materials like FA and CA. PC3 is strongly associated with curing periods and compressive strength, highlighting the impact of curing time on the strength of the mortar.

The clustering of the WTS replacement proportion, amount of WTS, and slump values in the same principal component indicates that these variables are closely related and exhibit similar patterns of variation within the dataset. This alignment suggests that the addition of WTS significantly influences the workability of the concrete mix, as reflected in the slump values. The consistent order of these variables in the principal component highlights their shared impact on the properties of the concrete mix, implying that changes in WTS proportion and amount directly affect the slump and overall workability.

4.6 LEACHING-INDUCED VARIATIONS IN HEAVY METAL CONCENTRATIONS

4.6.1 Properties of heavy metals in WTS and sand

The analysis of heavy metal concentrations in the WTS sludge and Sylhet sand reveals important insights into their potential environmental impact, particularly when compared to other materials used in construction. **Table 4.7** presents the concentrations of heavy metals found in WTS and Sylhet sand, alongside the reported concentrations for other supplementary materials used in construction from various studies. The WTS exhibits minimal concentrations of heavy metals, with Copper (Cu) recorded at 29.54 mg/kg, Chromium (Cr) at 31.37 mg/kg, Cadmium (Cd) at 11.59 mg/kg, Zinc (Zn) at 74.14 mg/kg, Nickel (Ni) at 23.45 mg/kg, and Iron (Fe) at 19.62 mg/kg. The concentrations of Cu, Cr, Cd, Mn, and Ni in WTS are 1.71–14.72, 1.38–13.82, 1.621–14.76, 2.02–19.02, and 1.25–5.95 times lower than in most supplementary materials reported by other researchers. However, the concentrations of Cr and Cd in WTS are higher than those found in tannery and textile sludge. The result suggests that while WTS generally contains lower heavy metal concentrations, certain metals like Cr and Cd may still be present at higher levels than other supplementary materials.

While each of these concentrations may seem insignificant alone, together they pose environmental and human health risks through bioaccumulation and bio-magnification if the sludge is not handled properly [29]. Sylhet sand, used as fine aggregate in concrete in this study, has slightly higher heavy metal concentrations compared to WTS. The Cu content (64.43 mg/kg) in Sylhet sand is more than double that of WTS, and Fe levels (68.55 mg/kg) are over three times higher than in WTS (19.62 mg/kg). Moreover, Cu is 1.76 times higher and Ni levels (40.56 mg/kg) are about 1.73 times higher in Sylhet sand than in WTS. These elevated concentrations could lead to significant environmental impacts, particularly through leaching from concrete into nearby water bodies, posing risks to aquatic ecosystems.

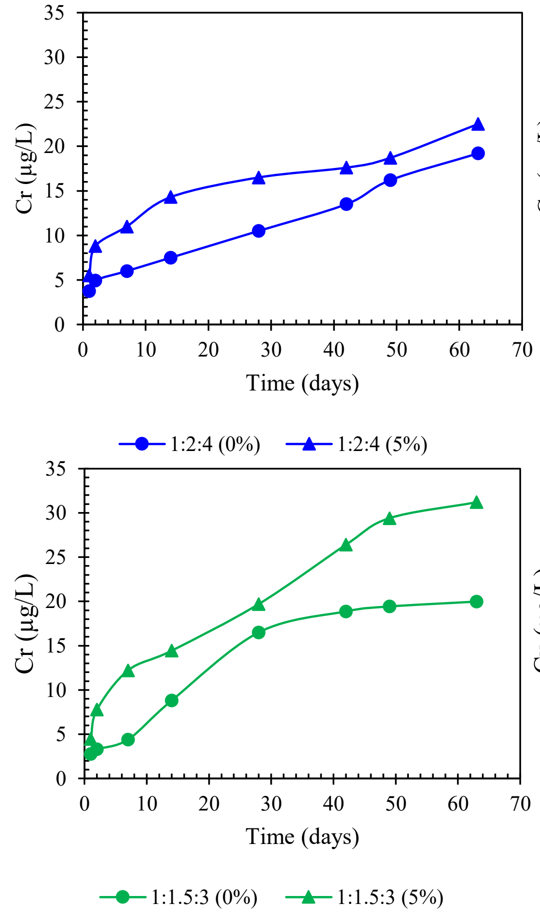
Table 4.7. Concentration of heavy metals found in the WTS and Sylhet sand.

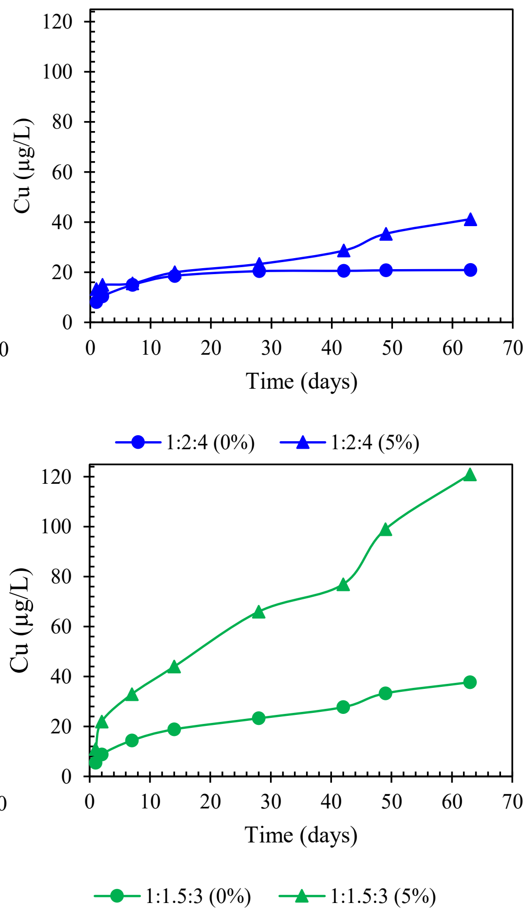
Concentrations (mg/kg)	Cu	Cr	Fe	Cd	Mn	Ni	Year	References
WTS	29.54	31.37	19.62	0.58	74.14	23.45	2024	This study
Sylhet Sand	64.43	15.14	68.55	1.79	7.98	40.56	2024	This study
Fly Ash	-	63.8-95.9	-	0.07-0.65	-	26.7-32.6	2022	[209]
Sewage sludge	370	73	-	2.17	460	-	2022	[210]
Solid Waste	434.7	433.5	-	8.56	-	-	2018	[211]
Textile sludge	58	10	-	5.6	-	32	2018	[212]
Tannery sludge	385.7	19.23	-	<0.01	-	139.5	2017	[59]
Sewage sludge	146.96	43.18	-	0.94	-	29.32	2013	[213]
Textile sludge	225.48	231.45	-	5.17	-	89.67	2009	[214]
Fly Ash	25-76	58-90	-	0.10-0.60	150-1410	-	2005	[215]

However, the heavy metal concentrations in Sylhet sand are still lower than those found in other supplementary construction materials used globally. For instance, Cr levels in Sylhet sand are 2–6 times lower than in fly ash, where Cr ranges from 63.8 to 95.9 mg/kg, depending on the specific samples.

4.6.2 Trend of heavy metal concentration due to leaching

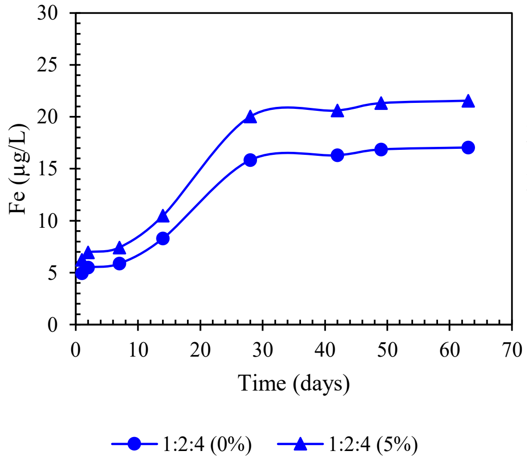
Figure 4.18 shows the long-term changes in the concentrations of heavy metals in concrete specimens with mix ratios of 1:2:4 and 1:1.5:3, where 5% of the cement was replaced by Water Treatment Sludge (WTS). The heavy metal concentrations follow a specific trend, with Copper (Cu) having the highest levels, ranging from 41 to 121 μg/L. This is followed by Manganese (Mn) with concentrations between 28 and 88 μg/L. Nickel (Ni) shows concentrations ranging from 31 to 33 μg/L, while Chromium (Cr) has levels between 23 and 31 μg/L. Iron (Fe) concentrations range from 22 to 28 μg/L, and Cadmium (Cd) is present in the lowest concentrations, ranging from 3 to 12 μg/L. The concentration of heavy metals in the concrete specimens shows a general increasing trend for all cases. Specifically, this upward trend is evident in concrete with 5% WTS replacement, with concentrations being 1.07 to 4.41 times higher in the 1:2:4 mix ratio and 1.04 to 4.14 times higher in the 1:1.5:3 mix ratio compared to their respective control concrete specimens.

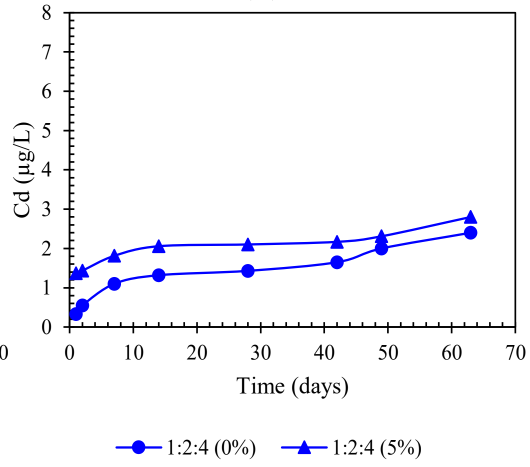




(a) Cr

(b) Cu





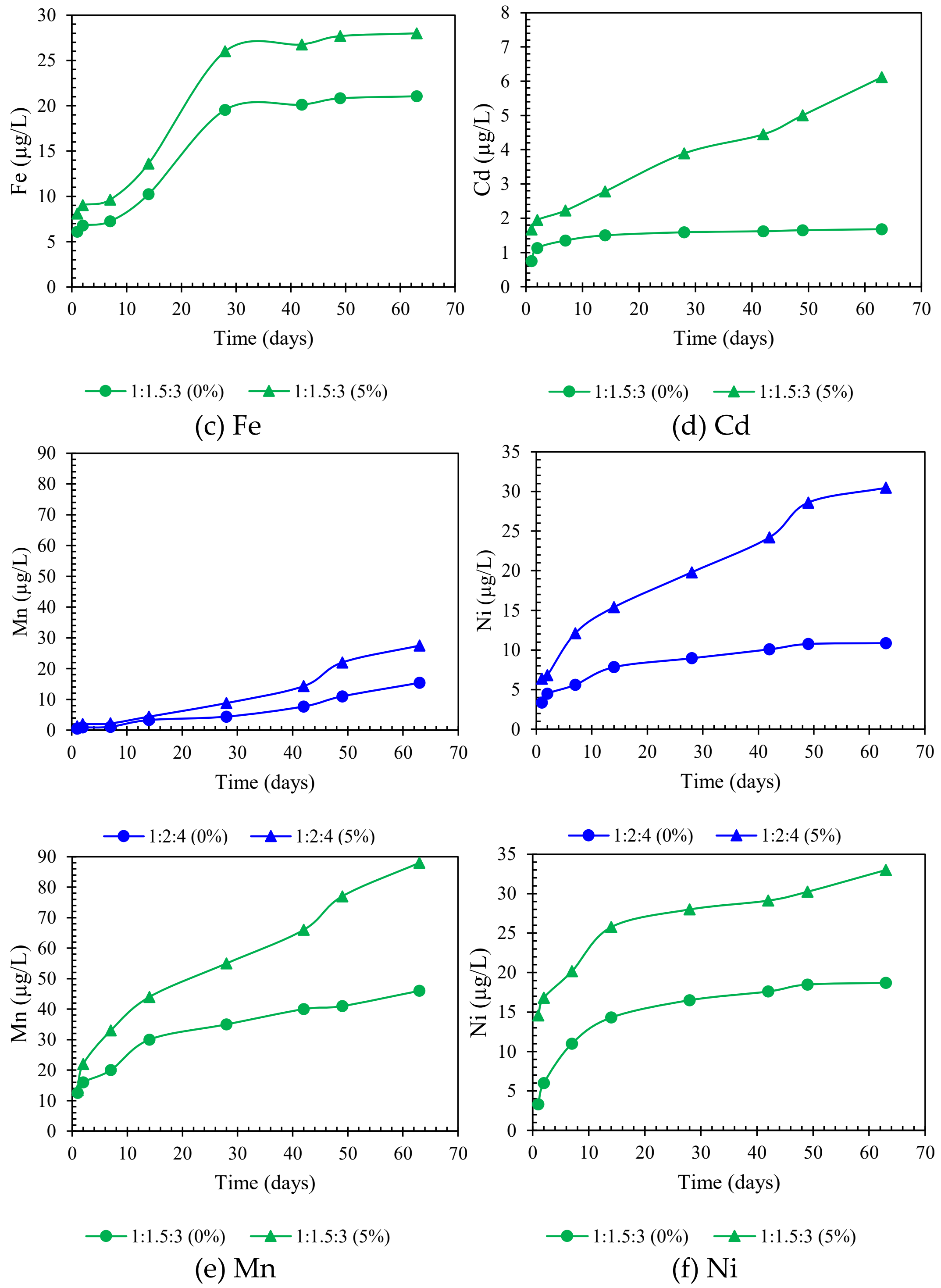


Fig. 4.18 : Leaching behavior of the heavy metals of the concrete specimen at 63 days leaching periods.

Moreover, the concentration levels of heavy metals in the 1:1.5:3 concrete mix ratio are notably higher than those in the 1:2:4 mix ratio. The concentrations are found to be 1.02 to 25 times greater for the control specimens, while the concentrations are 1.01 to 15 times higher for the 5% WTS replacement specimens. The higher concentrations of heavy metals in the 1:1.5:3 concrete mix ratio compared to the 1:2:4 mix ratio can be primarily attributed to the different proportions of cement and aggregate. In the 1:1.5:3 mix, with a higher cement content, the partial replacement of cement with WTS leads to a greater release of heavy metals. This is because the increased cement content enhances the interaction between the WTS and the concrete matrix [216], resulting in higher metal concentrations. Additionally, the lower water-to-cement ratio in the 1:1.5:3 mix generally makes the concrete denser [217,218], which can facilitate the release of heavy metals from the WTS. Furthermore, the concentration of heavy metals obtained from leaching extraction was substantially lower than the standard values set by the USEPA, as detailed in **Table 3.6**.

Furthermore, it is observed that the concentrations of heavy metals tend to stabilize after approximately 40 days. However, the concentration levels exhibit a marked increase before leveling off during the initial period, especially within the first 15 days. The observed stabilization of heavy metal concentrations after approximately 40 days, following a significant increase during the first 15 days, can be explained by the initial dynamics of the leaching process and concrete curing. In the early stages, the concrete is more porous, allowing for a higher rate of heavy metal leaching as the material is still curing and more permeable [219,220]. This leads to a marked rise in concentrations. However, as the concrete continues to cure, the formation of less permeable phases reduces porosity, thereby decreasing the leaching rate. Additionally, the readily leachable heavy metals become depleted over time, and protective layers may form on the concrete surface, further limiting the release of metals. This combination of factors results in the stabilization of heavy metal concentrations after the initial rapid increase[213,221,222].

Figure 4.19 indicates that after 63 days, all heavy metals, except for Nickel (Ni), have higher concentrations in the 1:1.5:3 concrete mix ratio compared to the

1:2:4 mix ratio. The differences in concentration between the two ratios are significant, with increases of 6.6% for Iron (Fe), 123.4% for Copper (Cu), 39.0% for Chromium (Cr), 274.4% for Cadmium (Cd), and 12.7% for Manganese (Mn). The concentrations of heavy metals in the concrete specimens with partial WTS replacement were observed to be significantly higher after 63 days of leaching. Specifically, for the 1:2:4 mix ratio, the increases were 26.4% for Iron (Fe), 97.2% for Copper (Cu), 17.2% for Chromium (Cr), 16.7% for Cadmium (Cd), 78.6% for Manganese (Mn), and 180.4% for Nickel (Ni) compared to the control specimen. In the 1:1.5:3 mix ratio, the increases were even more pronounced, with concentrations 33.0% higher for Fe, 220.6% for Cu, 56.2% for Cr, 264.0% for Cd, 91.3% for Mn, and 76.5% for Ni compared to the control specimen. Heavy metals leaching from concrete can originate from various sources including WTS and Sylhet sand. WTS may contribute metals due to chemical coagulants used in water treatment process [22,24]. In addition, WTS can contain residual metals from the raw water sources, which may include various natural and anthropogenic contaminants present before treatment [223,224].

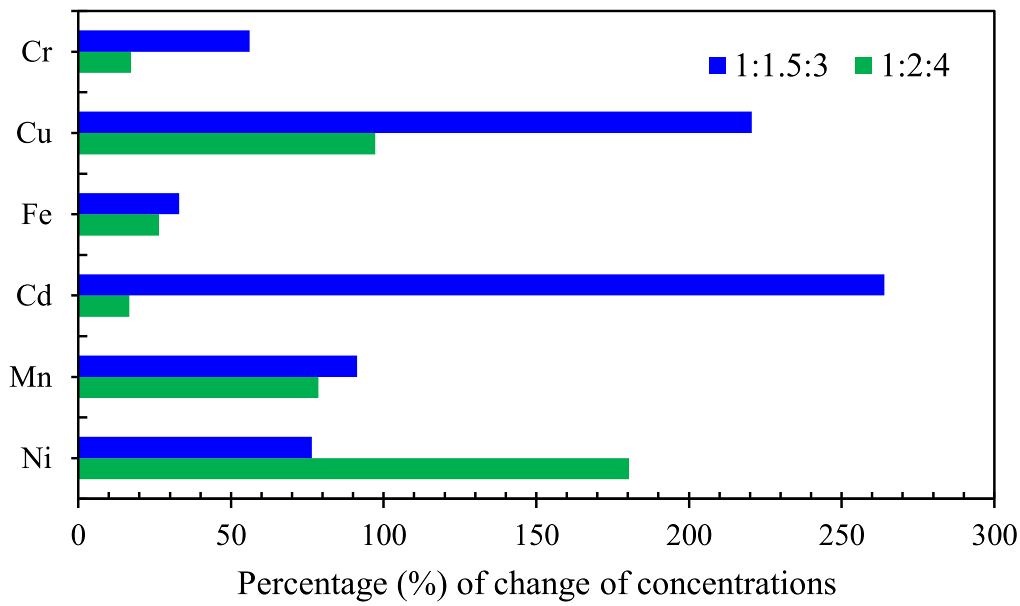


Fig. 4.19 : Variations in the concentration of heavy metals after partially replacing cement with WTS in concrete after 63 days.

Sylhet sand, collected from riverbeds, might naturally contain heavy metals from its mineral composition [225]. Leaching occurs when these metals dissolve and migrate through the concrete, driven by factors such as the alkaline pH of concrete, its porosity, and environmental conditions like rainfall and temperature [8,28,29]. Despite these potential sources, current data indicates that heavy metal concentrations and leaching are significantly low, underscoring the importance of rigorous monitoring and adherence to environmental regulations to ensure safety and mitigate risks.

4.6.3 Statistical Analysis of Changes in Heavy Metal Concentrations Due to Leaching

Table 4.8 illustrates how leachate concentrations of heavy metals—Chromium (Cr), Copper (Cu), Iron (Fe), Cadmium (Cd), Manganese (Mn), and Nickel (Ni)—change over 63 days for concrete specimens with and without 5% Water Treatment Sludge (WTS) replacement, across two concrete mix ratios (1:2:4 and 1:1.5:3). The addition of WTS generally increases the concentration of most metals in the 1:2:4 concrete mix ratio, notably Cu, Fe, Mn, and Ni, while Cr and Cd show varied trends, with Chromium decreasing and Cadmium decreasing slightly. Although the concentration of Cr and Cd in the concrete specimens with WTS replacement was higher compared to the control specimens, as shown in Figure 4.8.

Table 4.8. Trend of changes of leachate parameters over the 63 days for the control and 5% WTS replacement for concrete specimen.

Parameters	Ratio: 1:2:4				Ratio: 1:1.5:3			
	0%	Sig. Mark	5%	Sig. Mark	0%	Sig. Mark	5%	Sig. Mark
Cr	0.241	***	0.220	***	0.304	***	0.428	***
Cu	0.134	+	0.434	**	0.473	**	1.610	***
Fe	0.216	***	0.273	***	0.267	****	0.355	***
Cd	0.030	+	0.020	***	0.007	+	0.070	***
Mn	0.236	*	0.418	**	0.490	+	1.048	**
Ni	0.105	*	0.388	***	0.158	*	0.239	**

Note: Values were statistically significant at 90% (+), 95% (*), 99% (**), and 99.99 (***) confidence interval.

In contrast, the 1:1.5:3 mix ratio exhibits more significant increases across all metals, with Cu, Cd, and Mn showing significant rises, indicating that the higher cement content and different mix proportions enhance the leaching of these metals. The variations in leachate concentrations highlight the impact of WTS on heavy metal release, with the 1:1.5:3 mix ratio showing generally higher concentrations due to its composition and interactions between WTS and the concrete matrix. The observed changes in concentration, although significant, show a range of variation between 0.020 and 1.61 μg/L per day. This range suggests that it will require a prolonged period before the concentrations reach levels that exceed the regulatory standards. Furthermore, as depicted in Figure 4.18, the rate at which the concentration changes become stable after around 40 days. This stabilization indicates that the concentration will not rise dramatically beyond current levels, thus minimizing the likelihood of causing major environmental problems.

Table 4.9 presents the results of a Paired Sample T-test, which was conducted to assess whether there are significant differences in the average concentrations of heavy metals in leachate samples between control specimens and concrete specimens that incorporated Water Treatment Sludge (WTS) as a replacement material. This analysis was performed for both concrete mix ratios. Additionally, the test also evaluated differences between the two mix ratios for both control specimens and those with a 5% WTS replacement. The analysis was performed for two distinct concrete mix ratios, 1:1.5:3 and 1:2:4. For the 1:1.5:3 mix ratio, significant changes were observed in the concentrations of Cr, Mn, Ni, Cu, Cd, and Fe when WTS was introduced. The p-values for these metals were all below 0.05, indicating that the differences are statistically significant. For example, Ni showed a particularly strong reduction, with a t-value of -23.20 and a p-value of 0.000, highlighting the substantial impact of WTS on reducing metal concentrations.

Similarly, the addition of 5% WTS also led to significant decreases in the same heavy metals in the 1:2:4 mix ratio, with p-values consistently below 0.05. Notably, Copper and Cadmium both exhibited highly significant reductions, with identical t-values of -7.32 and p-values of 0.000, suggesting that WTS effectively changes these metal concentrations in the leachate. Overall, the results indicate that incorporating WTS in concrete significantly reduces the leachate concentrations of heavy metals, regardless of the mix ratio used.

Table 4.9 also highlights significant differences in heavy metal concentrations between the 1:1.5:3 and 1:2:4 concrete mix ratios for both control specimens (0% WTS) and those with a 5% WTS replacement. In the control specimens, the mix ratios resulted in significant changes in the concentrations of Mn, Ni, and Fe, with p-values well below 0.05, confirming the statistical significance of these differences. Both concrete mix ratios similarly led to significant differences in the concentrations of Mn, Ni, Cu, Cd, and Fe, with particularly notable increases in Mn and Ni for the 5% WTS replacement specimens. The findings suggest that the concrete mix ratios have a substantial impact on the heavy metal content in the leachate. However, some parameters, including Cr in both control and 5% WTS specimens, as well as Cu and Cd in the control specimens, did not show statistically significant differences and were not notably affected by the mix ratios.

Table 4.9. Results of Paired sample T-test for the leachate parameter (two tailed) at 95% confidence interval.

Paired variables	Leachate Parameter	Mean	Std. Deviation	t	Sig.	Sig. Mark
1: 1:1.5:3 (0%) and 1: 1:1.5:3 (5%)	Cr	-6.43	3.31	-5.49	0.001	*
	Mn	-19.74	14.25	-3.92	0.006	*
	Ni	-11.48	1.40	-23.20	0.000	*
	Cu	-2.08	1.35	-4.35	0.003	*
	Cd	-2.08	1.35	-4.35	0.003	*
	Fe	-4.63	2.31	-5.65	0.001	*
1:2:4 (0%)	Cr	-4.15	1.72	-6.82	0.000	*
	Mn	-4.76	4.68	-2.88	0.024	*
	Ni	-10.21	6.51	-4.44	0.003	*

and 1:2:4 (5%)	Cu	-0.66	0.26	-7.32	0.000	*
	Cd	-0.66	0.26	-7.32	0.000	*
	Fe	-2.99	1.51	-5.59	0.001	*
1:1.5:3 (0%) and 1:2:4 (0%)	Cr	1.55	3.05	1.44	0.193	
	Mn	24.53	7.98	8.69	0.000	*
	Ni	5.48	3.08	5.02	0.002	*
	Cu	4.36	7.12	1.73	0.127	
	Cd	0.08	0.42	0.51	0.626	
	Fe	2.65	1.33	5.65	0.001	*
1:1.5:3 (5%) and 1: 2:4 (5%)	Cr	3.83	4.85	2.23	0.061	
	Mn	39.50	17.22	6.49	0.000	*
	Ni	6.74	3.34	5.71	0.001	*
	Cu	35.08	28.44	3.49	0.010	
	Cd	1.49	1.17	3.60	0.009	
	Fe	4.29	2.13	5.69	0.001	*

Figure 4.20 illustrates the relationship between the heavy metals extracted during the leaching process from the control concrete specimen and the concrete specimen with a 5% WTS replacement in cement. The significant positive correlation of the leaching period with all heavy metals except Mn and Ni for the control specimen suggests that the duration of leaching strongly influences the extraction of most heavy metals, but has little to no effect on the leaching of Mn and Ni. The trend analysis, as mentioned in **Table 4.7**, also shows that Mn and Ni exhibit comparatively less significant trends.

In contrast, the concrete specimen with a 5% WTS replacement shows a stronger correlation between the leaching period and heavy metal concentrations compared to the control concrete specimen, as indicated by higher correlation coefficient values. The results indicate that replacing 5% of cement with WTS in concrete enhances the correlation between the leaching period and heavy metal concentrations, suggesting that WTS influences the leaching behavior of heavy metals more significantly than in control concrete. Despite this, the correlation for Mn remains positive but not statistically significant, even though its correlation coefficient increases from 0.53 to 0.65. The trend analysis as mentioned in **Table 4.7**, which also shows a rise in the significance level from 90-95% to 99% confidence interval for Mn, suggests that while the relationship is strengthening, it is still not significant enough to conclusively impact Mn leaching behavior.

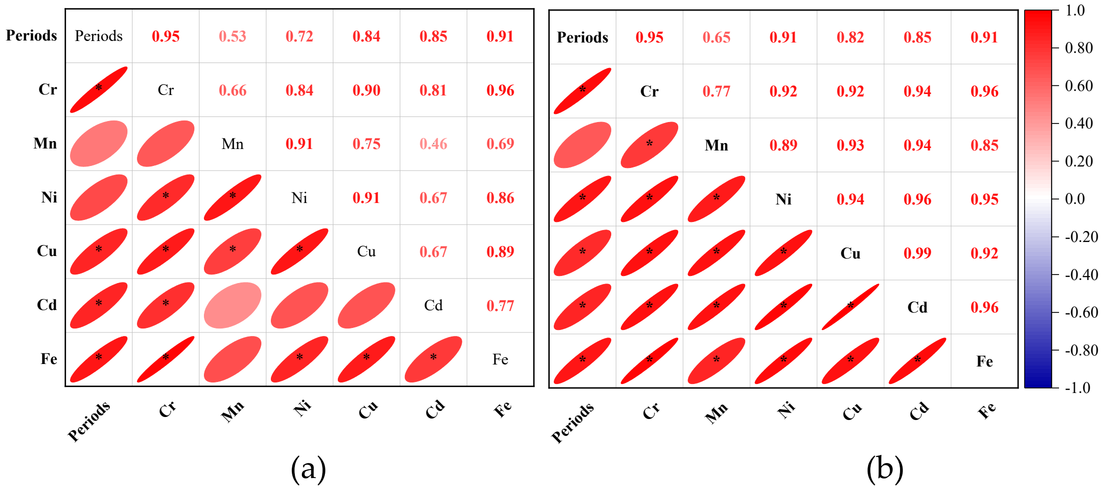


Fig. 4.20 : Correlation plot of heavy metals in (a) control and, (b) 5% WTS replacement concrete specimens.

All heavy metals exhibit a significant positive correlation with each other in both cases which means that the leaching of one heavy metal is likely to influence or be associated with the leaching of others. The factors affecting the leaching of heavy metals, such as the composition and characteristics of the concrete and sludge, may influence the leaching of multiple metals simultaneously. Cd shows a more significant correlation with most heavy metals in the concrete specimen with 5% WTS replacement, indicating that its leaching behavior is more closely related to the leaching of other metals compared to the control specimen. This contrasts with the control concrete, where Cd only demonstrates a significant relationship with Cr and lacks significant correlations with most other heavy metals. The increased number of significant correlations in the WTS-replaced specimen suggests that the presence of WTS influences the interactions between Cd and other heavy metals, leading to a more interconnected leaching behavior.

4.7 LIFE CYCLE IMPACTS ON ENVIRONMENT FOR CEMENT PRODUCTION

Table 4.10 provides a detailed comparison of environmental impacts associated with cement production when clinker is replaced with water treatment plant sludge (WTS). The most significant reduction is observed in the category of climate change, where the carbon footprint decreases by 1.91%, translating to a reduction from 2275.44 kg CO_2-Eq to 2231.98 kg CO_2-Eq for 5% WTS replacement level. This reduction highlights the potential of WTS to lower greenhouse gas emissions, a crucial factor in addressing global warming and mitigating climate change impacts. The use of WTS thus demonstrates a tangible benefit in reducing the carbon footprint of cement production. The carbon footprint value was notably higher than those reported by Stafford et al. (2016) [138], Tun, Bonnet, and Gheewala (2020) [226], and Thwe, Khatiwada, and Gasparatos (2021) [140], which were 1113 kg CO_2-Eq, 632 kg CO_2-Eq, 872-1150 kg CO_2-Eq, respectively for 1000 kg of cement production. Although, Wolde et al. (2024) [139] reported a carbon footprint value of 4045 kg CO_2-Eq, which is substantially higher than the value observed in this study. This discrepancy +can be explained by the different assumptions and life cycle stages considered in various studies, with many of them focusing primarily on the Gate-to-Gate stage.

Table 4.10. Environmental impacts associated with cement production on mid-point impact categories for 1000 kg of cement production.

Impact categories	Units	0%	2.5%	5.0%	7.5%	10%	15%
Agricultural land occupation	m^2a	48.99	48.90	48.80	48.71	48.61	48.42
Climate change	kg CO_2-Eq	2275.44	2253.71	2231.98	2210.24	2188.51	2145.05
Fossil depletion	kg oil-Eq	359.04	356.27	353.50	350.73	347.96	342.42
Freshwater Ecotoxicity	kg 1,4-DCB-Eq	157.10	157.03	156.96	156.89	156.82	156.69
Freshwater Eutrophication	kg P-Eq	0.581	0.578	0.575	0.571	0.568	0.561
Human toxicity	kg 1,4-DCB-Eq	2106.95	2104.06	2101.17	2098.27	2095.38	2089.59
Ionising radiation	kg U_{235}-Eq	38.61	38.39	38.17	37.96	37.74	37.30
Marine ecotoxicity	kg 1,4-DCB-Eq	141.07	141.00	140.93	140.87	140.80	140.67
Marine eutrophication	kg N-Eq	4.15	4.13	4.11	4.08	4.06	4.02
Metal depletion	kg Fe-Eq	23.02	22.87	22.73	22.58	22.44	22.15
Natural land transformation	m^2	0.084	0.083	0.081	0.080	0.079	0.076
Ozone depletion	kg CFC-11-Eq	0.000	3.6E-05	3.6E-05	3.5E-05	3.5E-05	3.4E-05

Impact categories	Units	0%	2.5%	5.0%	7.5%	10%	15%
Particulate matter formation	kg PM_{10}-Eq	4.901	4.88	4.86	4.83	4.81	4.77
Photochemical oxidant formation	kg NMVOC	6.057	6.00	5.94	5.88	5.82	5.70
Terrestrial acidification	kg SO_2-Eq	4.964	4.91	4.86	4.80	4.75	4.65
Terrestrial ecotoxicity	kg 1,4-DCB-Eq	3.860	3.86	3.86	3.86	3.86	3.86
Urban land occupation	m^2a	55.673	54.75	53.83	52.91	51.98	50.14
Water depletion	m^3	1.419	1.41	1.41	1.40	1.40	1.39

In addition to climate change, several other impact categories show notable reductions, when WTS is used instead of clinker. Other impact categories demonstrate a reduction ranging from 0.09% to 3.31% for 5% WTS replacement level in concrete. These categories include Natural Land Transformation, Urban Land Occupation, Ozone Depletion, Terrestrial Acidification, and Photochemical Oxidant Formation. These reductions indicate that using WTS in cement production not only conserves fossil resources but also minimizes the adverse effects on land use and atmospheric conditions. Such improvements reflect a positive shift towards more sustainable cement production practices. Conversely, the reduction rates for Freshwater Ecotoxicity, Marine Ecotoxicity, Human Toxicity, Particulate Matter Formation, Terrestrial Ecotoxicity, and Water Depletion are notably lower.

Figure 4.21 represents the contribution of each life cycle stage to mid-point environmental impact categories for 5% WTS replacement level. The Cradle to Gate stage stands out as the dominant phase for 39% of the impact categories in the lifecycle assessment, emphasizing the substantial environmental impacts associated with resource extraction. The result indicates that the extraction of raw materials significantly impacts the environment, as this phase involves activities such as the use of explosives, fuel combustion, and various fugitive emissions [226]. On the other hand, around 33% of the impact categories reveal that the Gate to Grave stage significantly contributes to the overall impact which suggests that end-of-life processes, such as disposal and treatment of products. This is attributed to the uncontrolled generation rate and inadequate management of construction and demolition waste in Bangladesh [227,228]. Furthermore, the remaining 28% of impact categories show that the Gate-to-Gate stage, which encompasses manufacturing processes, is also a significant contributor to environmental impacts.

Cement manufacturing processes depend largely on electricity in Bangladesh, with 26%, 7.14%, and 2.73% of it coming from furnace oil, diesel, and coal powered electrical plant, respectively [229]. These power plants significantly contribute to environmental pollution [230,231]. The result highlights the importance of considering the impacts associated with the production phase in the life cycle assessment. **Figure 4.21** also indicates that impact categories such as Climate Change and Freshwater Eutrophication are significantly affected by multiple stages of the product life cycle.

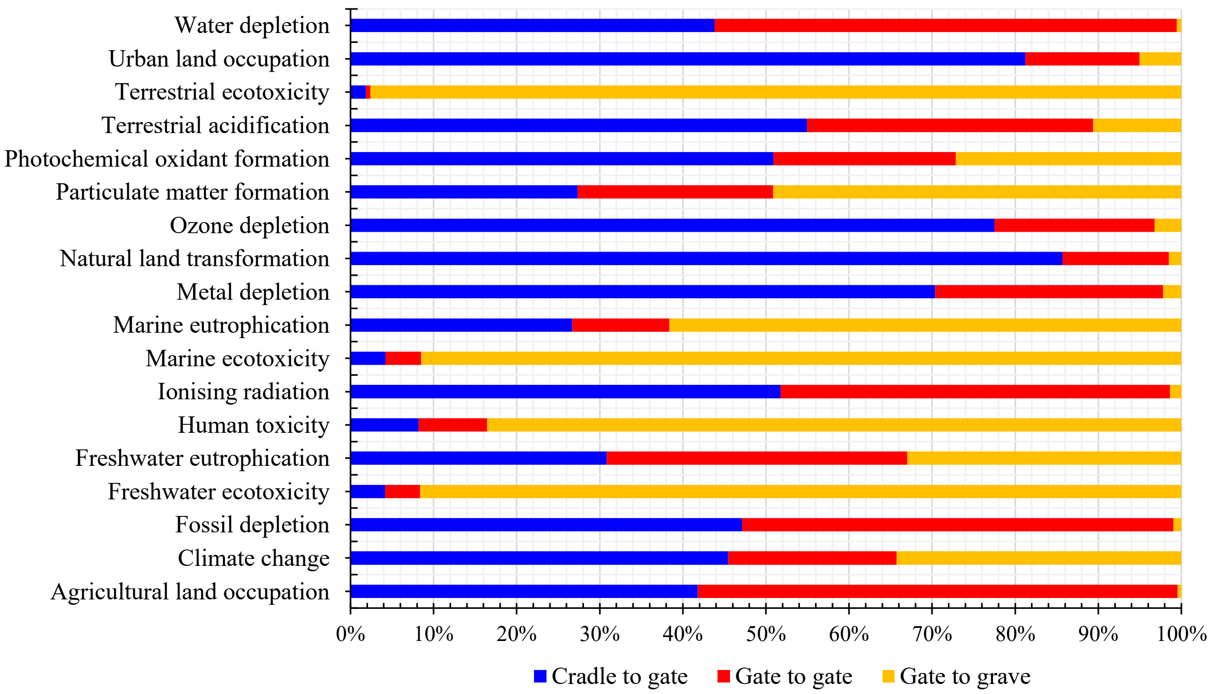


Fig. 4.21 : Proportion of mid-point environmental impact categories contributed by each life cycle stage for 5% WTS replacement level in concrete.

Climate Change impacts are notably contributed to by both the Cradle to Gate phase (46%) and the Gate to Grave phase (34%), which signifies that both the production and end-of-life stages are key contributors to these environmental effects. However, Freshwater Eutrophication shows a more balanced distribution of impacts across the Cradle to Gate (31%), Gate to Gate (36%), and Gate to Grave (33%) stages. This balance highlights the need for targeted interventions throughout the entire life cycle to address the issue effectively. In contrast, categories like Freshwater Ecotoxicity and Human Toxicity are predominantly influenced by the Gate to Grave stage (84-92%), underlining the importance of focusing on the disposal phase to mitigate these effects. On the other hand, Fossil Depletion and Agricultural Land Occupation are primarily driven by the Cradle to Gate and Gate to Gate stages, indicating a need for improvements in production and operational processes. This varied distribution of impacts underscores the necessity for a holistic approach to environmental management, one that addresses issues at specific stages of the life cycle.

4.8 SUMMARY

The study demonstrates that incorporating up to 5% WTS as a partial cement replacement significantly enhances concrete workability, with slump values improving to between 110-145 mm. This improvement is due to WTS's larger and irregular particles, which facilitate better mixing and placement. Compressive strength is optimized at a 5% WTS replacement, providing stronger concrete compared to higher levels of substitution, which begin to reduce strength. The use of WTS also results in a decrease in unit weight for both concrete and mortar, making them lighter, though this reduction can negatively impact compressive strength at higher replacement rates. Heavy metal leaching is a notable concern, with increased levels of Cu and Mn observed in concrete containing WTS, although overall leaching remains within regulatory limits. The study emphasizes the importance of monitoring leaching behavior to manage environmental risks. Life cycle assessment (LCA) results highlight that replacing 5% of clinker with WTS in cement production reduces the carbon footprint by 1.91% and mitigates other environmental impacts by 0.09% to 3.31%. This reduction in environmental impact underscores WTS's potential as a sustainable and environmentally friendly alternative in the construction industry, offering both material and ecological benefits.

Chapter 5: CONCLUSIONS

5.1 GENERAL

The chapter explores the potential of water treatment plant sludge (WTS) as a sustainable partial replacement for cement in concrete and mortar. The study found that incorporating WTS enhances workability and compressive strength, particularly at 5% replacement levels, due to its pozzolanic activity. However, higher WTS levels reduce strength and increase heavy metal leaching, although within regulatory limits. A life cycle assessment (LCA) indicates a reduction in environmental impact when WTS is used. The study underscores the need for careful integration of WTS in construction, balancing sustainability with structural performance, and recommends further research to address long-term durability and environmental safety.

5.2 KEY FINDINGS

The key outcomes of this study are given as follows:

- The WTS particles were found to be mostly irregular in shape, with an average size of 9.20 μm, which is larger than the 3.88 μm average particle size of OPC. The high content of SiO_2 in WTS (45.40%), along with notable amounts of Al_2O_3 and Fe_2O_3, met the standards for pozzolanic activity, making WTS a viable supplementary cementitious material.
- The inclusion of WTS in concrete mixes enhanced workability, with slump values ranging from 110 mm to 145 mm, particularly with up to 5% WTS replacement. Moreover, compressive strength of concrete peaked at 5% WTS replacement. A 1:2 mix ratio cement mortar with 7.5% WTS replacement exhibited the highest compressive strength. However, compressive strength

declined at higher WTS replacement levels. The inclusion of WTS in concrete reduces its unit weight compared to OPC, leading to a lower overall density. This reduction in weight came with a trade-off, as higher WTS replacement levels led to diminished compressive strength.

- The study found that 5% WTS replacement in concrete increased heavy metal leaching, particularly Cu and Mn, but concentrations stabilized after 40 days due to reduced porosity and protective layer formation. While the 1:1.5:3 mix showed higher heavy metal levels than the 1:2:4 mix, overall leaching remained within safe limits, ensuring environmental safety with proper monitoring. However, LCA showed that 5% WTS use in cement reduced the carbon footprint by 1.91% and mitigated environmental impacts by up to 3.31%.

5.3 IMPLECATIONS OF THE STUDY

The findings of this study underscore the significant potential of incorporating WTS as a partial replacement for cement in concrete and mortar, highlighting its contributions to sustainable construction practices WTS serves as an effective supplementary cementitious material by improving workability and reducing the carbon footprint that not only enhances the performance of construction materials up to certain replacement levels but also promotes environmental conservation through resource reuse and greenhouse gas reduction. The demonstrated pozzolanic activity of WTS, evidenced by its chemical composition, contributes to the strength and durability of the mixes, while the observed reduction in unit weight offers benefits for specific structural applications where lighter materials are advantageous. However, the study also reveals critical considerations regarding the optimal replacement levels, as exceeding these limits can lead to diminished mechanical properties and increased heavy metal leaching, albeit within regulatory confines. The strong correlations between WTS proportions and both material performance and environmental impacts emphasize the need for careful integration of WTS into construction practices to balance sustainability with structural integrity.

5.4 RECOMMENDATIONS

Based on the outcomes of the study, it is recommended that the construction industry incorporate WTS as a partial cement replacement at around 5% for improved workability and strength, while minimizing environmental impact. Monitoring and adhering to regulatory standards for heavy metal leaching is crucial, especially for long-term durability and moisture exposure. Tailored mix designs should be developed to suit specific construction needs. Further research is needed to assess the long-term durability of WTS-modified concrete and investigate methods to mitigate heavy metal leaching. Implementing these recommendations can promote sustainable WTS adoption in construction, reducing environmental impact and supporting responsible building practices.

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