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Developing an eco-friendly cementitious grout using paper sludge ash and steel fiber recovered from waste tires

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A B S T R A C T

Cement is the most important component in cementitious grout production,andtt accounts for 7% of global greenhouse gas emissions. Reducing the amount of cement used in infrastructure and buildings is a desirable way to lower the total carbon footprint associated with grout production. relatedly, developments in manufacturing and transportation lead to the production of cars in large numbers, which in turn leads to an increase in the production of byproduct waste like waste tires, which are thrown away directly without recycling. Thus, this study aims to develop a novel sustainable grout by recycling paper sludge ash waste (PSA) and waste steel fiber (WSF) as partial cement replacements. This will help reduce grout production costs, minimize environmental pollution during cement production, and enhance landfill and waste management. The Grout mixtures used in this paper were prepared using ordinary Portland cement (OPC), WSF extracted from vehicle tires, PSA, and water. Different proportions of WSF(0, 1, 2, 3%) and PSA(5, 10%) were used in the weight of cement in designing sustainable grout. The mechanical properties of the sustainable grout were evaluated by examining extensive tests including flow tables, compressive strength, and flexural strength. The results showed that partially replacing the cement with 3% WSF and 1% WSP with 5% PSA resulted in a significant improvement in workability ,as well as a clear increase in compressive and flexural strength at an early age compared to the reference mixture.

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1. Introduction

Improvements in technology have a substantial impact on economic growth and the development of many economic sectors, notably industrial ones [1, 2]. However, this improvement has resulted in an increase in environmental contamination rates and the development of new environmental concerns on a global scale [3]. Technological progress can accelerate the exploitation of natural resources such as minerals, fuels, and water [4]. Unsustainable resource extraction practices can lead to resource depletion, habitat destruction, and ecosystem loss, further exacerbating environmental challenges[5]. Consequently, governments and international health organizations have heightened their focus on addressing these

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environmental issues[6]. The demand for cement in various building and road construction projects has increased in recent years. A ton of Portland cement requires approximately 2.8 tons of natural resources, fuel, and other components. Cement manufacturers account for 8% of worldwide carbon dioxide emissions, with 40% coming from fuel burning and the rest from the limestone calcination process [7, 8]. Smoke from cement factories emits a large amount of carbon dioxide into the environment. It enters the atmosphere and forms granules that hinder sunlight from reaching the ground, resulting in forest fires and global warming[9]. Cement production accounts for almost seven percent of global greenhouse gas emissions[10].In order to reduce carbon dioxide emissions and environmental pollution, the world is currently moving toward using sustainable materials produced by industries or trash as renewable, accessible, and low-cost resources to partially replace cement. [11, 12]. Cement plays a very important role in grouting techniques, as it constitutes 80-90% of the components of injection materials. In spite of that, the grouting process is a common technique in many fields of civil engineering, such as construction, infrastructure repair, soil stabilization, crack filling, and soil improvement, especially dam foundations, but it has recently been used to fill the voids in porous asphalt pavement [13]. Cement plays a vital role in grout performance. Typically, a grouting process uses ordinary Portland cement (OPC) as a conventional grout material. However, researchers have investigated a variety of alternative materials to partially or completely replace OPC as grout. In the majority of cases, the replacement aims to reduce the cost of pavement construction and/or produce sustainable grout materials by recycling waste materials, which reduces emissions from the cement industry and reduces the cost of its demand [14, 15]. Many different types of building and ground improvement operations have long used grout. However, the composition, durability, and environmental effects of traditional grouts are often in need of improvement. Recently, there has been a growing focus on investigating more sustainable and eco-friendly alternatives[16].

2. Literature Review

Sustainability is the foundation of today's world of engineering. It can be described as using natural or by-product materials without influencing the future generation's demand, so using waste instead of Portland cement contributes to reducing environmental pollution[17]. Thus, in the last decade, road agencies, corporations, governments, and research institutes have given more attention to sustainability. Studies should typically concentrate on three components: economic advantages, social growth, and environmental conservation in the case of constructing sustainable pavement [18] A sustainable pavement (see Figure1-b) can be described as a safe, effective, ecosystem-healthy, open-space, and environmentally friendly pavement that meets essential needs efficiently uses resources and preserves or restores ecosystems [19].

Cement is a significant contributor to environmental pollution. There are several cement plants in Iraq, including Basra, Kufa, Muthanna, Kirkuk, Babel, and Karbala. The rise in demand for cement plants leads to elevated levels of $CO₂$ emissions, which disperse in the atmosphere as particles, obstructing sunlight and contributing to global warming [20]. Using waste material as a partial replacement for cement in the grouting technology that has been applied to civil engineering applications is beneficial and environmentally friendly and can contribute to minimizing global warming and reducing the cost of cement[21].

Bibliometrics, or analytical, is the study of collections of connected

materials, such as scientific literature. It provides well-established methodologies for assessing and showing links between research subjects, individuals, affiliations, or periodicals [22]. Numerous applications are compatible with the bibliometric approach, including Histcite, CiteSpace, Bib Excel, Vos Viewer, Pajek, and Gephi. This research created bibliometric and visual maps using a VOS viewer. Bibliometric network research is the primary use case for this program's single-architecture clustering and mapping capabilities. Network visualizations, density visualization tools, and overlay visualizations are the three types of visualizations that VOS viewers can generate [23]. Keywords cooccurrence analysis, which employs brief and visual maps, is among the most helpful ways to demonstrate the evolution of scientific topics and patterns [24]. The agreed-upon rules reveal the most crucial part of the picture: the maximum number of people: where each limitation represents an item. The Web of Science has more than 1000 keywords related to grouting materials. Previous studies and research (Figure 2) have commonly used the following keywords: compressive strength, cementitious grout, concrete, mechanical features, grout application, cement slurry, water absorption, durability, workability, and curing conditions. The combined author keywords, which span twelve cluster networks and amount to thirty items, are shown in Figures 3 and 4. As the number of nodes grows, it becomes clear how much weight keywords have in online searches. The most important studies relating to the design of cementitious grout can be found at these nodes; they include topics such as recycling, steel fiber, paper sludge ash, industrial waste, sustainability, cement replacement, strength development, and Portland cement. The research identified "supplementary cementitious materials" or "compressive strength " as the most crucial term. Furthermore, it seemed to have the longest chain and the most links when compared to other words. From most strongly bound to least, each distinct color represents a different sentence [25].

This analysis is used as an effective and useful tool in obtaining the most important related keywords in the current study and thus obtaining most of the previous studies related to the research topic. Similar to the combination and link visualizations, the item intensity visualization displays items according to their identifiers. In the item intensity depiction, the color at each position represents the item's strength. In the item intensity visualization, each location's color indicates the item's strength. Standard color schemes consist of blue, green, and yellow. Points tend to resemble yellow more in hue as their block size increases and the weights of the things blocking them increase. Not only is the opposite true, but it also holds that a point with fewer objects and lower object weights is one whose hue is closest to blue. A test helps visualize item strength[26-28]. Figure 5 displays the density visualization map of the (co-occurrences) in yellow color. Some of the most concentrated keywords and phrases with strong correlations include supplementary cementitious materials, compressive strength, and sustainability The researcher may find a diverse range of published papers using each of these keywords. The most common keywords are represented by large nodes, and the distance between them is depicted by a line since several keywords are used in the same search. Researchers have conducted numerous studies to develop sustainable grout materials. Most of these studies were devoted to reusing waste materials or by-product materials to replace the OPC as conventional grout material. Several waste materials were used to produce gout material, such as glass waste powder (GWP) [29], ceramic waste powder (CWP) [30], silica fume (SF), palm frond waste ash (PFWA)[31], and grout containing regular waste polyethylene terephthalate (PET) [32], etc.

The use of recycled paper as an input in the production of paper goods is on the rise around the globe. The papermaking process produces a substantial quantity of trash throughout the deinking and water treatment phases. Recent UK figures indicate an annual production of 4.5 million tons of paper, with 73% of the fiber originating from recovered paper. Different kinds of paper use this material [33]. One kind of pulp and paper industry waste known as paper sludge ash (PSA) is problematic from an economic and environmental perspective. However, researchers have recently investigated its possibilities as a sustainable and environmentally friendly material for various technological uses [34, 35]. Paper sludge ash was used in pavement design, construction, and concrete structures as a supplementary cementitious material. In concrete structures, wastepaper sludge ash (PSA) was a supplementary cement material to partially replace OPC [36]. The results showed that the use of PSA can lead to concrete performing better, in addition to being considered solid waste management. Devi et al.[37] investigated the use of PSA as a partial cement substitute in concrete. The researchers examined the impact of incorporating varying percentages of PSA (2.5%, 5%, and 7.5 % PSA) by weight of cement in concrete. The researchers conducted extensive tests to measure behavior, such as compressive strength, split tensile strength, and flexural strength, at 28 days of curing. The results of their study showed that the strength increased with the addition of PSA, up to 5 % of the replacement cement. On the other hand, waste rubber tires from vehicles are one of the most important environmental risks around the world. Hunyak et al. [38] investigated the effect of using wastepaper sludge ash (PSA) as a partial replacement for cement at 5% and 10% in concrete production. The study's findings show that adding PSA as an extra cementitious material makes the concrete stronger in flexural strength and creates a denser microstructure. The study's findings concluded that using

PSA may be a more environmentally friendly option than using

Waste paper sludge ash shows promise as a valuable resource in the construction industry, particularly for producing concrete, bricks, mortar, soil stabilizers, concrete pavements, and controlled low-strength materials[39]. Due to increased vehicle production, there is a need for proper disposal of large quantities of used rubber tires [40]. Therefore, researchers are exploring the potential applications of used tire rubber in construction technology.

Zia et al. [41] examined the usability of using a high dosage of steel fiber recovered from waste tire (WSF) in concrete. The researchers examined the impact of incorporating varying percentages of WSF (1% to 1.75% WSF) by weight of cement in concrete. The results of their study showed that adding WSF with a high content improves split tensile strength by 10% and flexural strength by 4%, as well as a 15% reduction in linear shrinkage in all of these percentages compared to normal concrete. Their study concluded that using WSF provides a sustainable solution to reduce the cost of cement and enhance the sustainability of construction and paving materials as an environmentally friendly option by reducing landfills. Firdawok et al. (2023) [42] used recycled steel fibers extracted from old vehicle tires (1, 1.5, 2, 2.5, and 3%) as a partial replacement of cement in reinforced concrete used in rigid pavement.

The study's findings revealed that 1.5% of steel fibers had a maximum compressive strength of 43.85 MPa compared to the reference mixture. While 3% of steel fibers had a maximum tensile strength of 4.75 MPa, the results of their study recommended the use of recycled steel fibers because they improve resistance to compressive, tensile, and flexural strength, as well as being a promising option for developing sustainable pavements. Based on available literature very limited studies investigate the effect of recycled paper sludge ash and waste fiber in modifying conventional grout. Therefore, this study aims to develop a sustainable cementitious grout material using recycled paper sludge ash and waste fiber as a partial replacement for cement.

Figure 1. Sustainability in civil engineering applications :(a) structure engineering, (b) pavement engineering [18] .

conventional cement.

Figure 2. Keywords network map 1 of using grout in civil engineering applications.

Figure 3. Keywords network map 2 of using grout in civil engineering

Figure 4. Keywords network map of using grout with waste materials in this study.

Figure 5. Keywords density visualization map.

Therefore, recycling this waste reduces the need for cement and its cost, provides an effective and vital option for disposal of this waste, helps in developing paving properties, and provides a sustainable solution to preserve the environment by reducing landfills for this waste. The cement grout specimens underwent comprehensive analysis to assess their physical and mechanical properties. Through rigorous testing, the optimal compositions of grouts were identified and refined. This examination aimed to measure the effectiveness and durability of the modified sustainable cement grouts in comparison to conventional cement-based grouts.

3. Research Methodology

To achieve the main aim of this research, the methodology involves developing grout mixtures through three phases. The flowchart in Figure 6 displays the research methodology, which involves the following stages:

- 1. The series I consists of four mixtures, comprising a control cement mix in addition to cement and waste steel fibers (WSF) with different proportions.
- 2. The Series II is to replace ordinary Portland cement (OPC) with supplementary cementitious materials of paper sludge ash (PSA) by 5% and 10%.
- 3. The Series III consists of the optimum proportion of waste steel fiber extracted from the first phase with cement and 5% and 10% of paper sludge ash.
- 4. The W/B ratio is constant at all phases and for all mixtures by 0.4%.

4. Raw Materials

Locally available cement was used in this research, while WSF and PSA were prepared in the laboratory. The materials included in the design of the grout materials are summarized as follows:

Commercial OPC CEM I 42.5R compatible with the Iraqi Standard No: 5/2019 [43] type I, which is used as a binder in all mixtures, is supplied by Saman Cement Factory. PSA is available in large quantities in the environment and thus it should be recycled to be used in various construction applications and reduce the accumulation of their size in the environment [44, 45].

At first, large quantities of paper were collected and burned in an open place at a temperature ranging from 350-450 °C as a primary burning to reduce its size, expel the oxides, and turn it into ash. The second stage is to burn the ashes in an electric furnace at a temperature of 800 °C for two hours [46]. The final stage is to grind the ash by a mill for thirty minutes to turn it into fine materials. The surface area specified using the Blaine method as per ASTM C204-11 [47] of OPC and PSA was 339 m²/kg and 1517 m²/kg, respectively. The bulk density of OPC was 2.74 g/cm³ and PSA was 2.63 g/cm³. The chemical properties of the OPC and PSA, clarify the main components as oxides specified by XRF appear in Table 1. WSF was prepared by collecting waste tires of damaged vehicles, then the fibers were extracted by machines. The fibers are cut to a length of 10 mm, with a diameter of 1 mm, and a density of 7700 kg/m^3 . Figure 7 displays the raw materials of sustainable cement-based grout.

Figure 6. The flowchart presents the methodology of the current investigation.

Figure 7. components of sustainable grout material: (a) water, (b) cement, (c) paper sludge ash, and (c) waste tire steel fiber

Grout materials samples used for mechanical characteristics measurement were prepared with a constant water-to-binder ratio of about 0.4%. Series I of designing the grout was by adding WSF to the mixture by 1%, 2%, and 3% by volume. Series II is the addition of PSA in different proportions of

5. Proportions of Grout Mixtures

cement 5% and 10% by weight.

While the series III adds the optimum percentage of WSF at 1% by volume with different proportions of PSA replacing 5% and 10% by weight with cement. The mixture grout proportions are described in Table 2. The dry materials represented by PSA were blended with OPC to the point of homogeneity, then these materials were added with water and blended in a mechanical mixer for 120 seconds according to ASTM C305-14 [48]. After the completion of mixing the dry materials with water, the WSF is gradually added to the homogeneous mixture and mixed well for a short period until homogeneity. After preparing the fresh grout sample, it is tested for flowability by the flow table according to the ASTM C230-14 [49].Then the fresh grout dough is poured into standard cubes with dimensions of 50 \times 50 \times 50 mm³ according to the ASTM C109-13 [50] for measuring the compressive strength at ages 7 and 28 days. In addition, the fresh grout is also poured into prisms with dimensions of $40\times40\times160$ mm³ according to the ASTM C348-14 [51] for measuring flexural strength at ages 7 and 28 days. After 24 hours of pouring grout into the molds, the molds are opened and the specimens are placed in the water basin until the required test day (Figure 8). The methods of testing in the current study are shown in Figure 9.

Table 1. The chemical characteristics of OPC and PSA

Oxides, $%$	OPC	PSA
SiO ₂	21.52	18.32
Al_2O_3	04.39	11.16
Fe ₂ O ₃	03.11	00.28
CaO	62.14	57.24
MgO	03.42	03.05
SO ₃	02.31	00.58
K,O	00.51	00.41
Na2O3	00.20	00.16

Figure 8. The cementitious grout preparation

Table 2. Components of grout mixtures

Sample	Series	OPC (gm)	PSA (gm)	WSF (gm)	Water (gm)
C		3700	000	000.0	1480
1WSF	I	3700	000	142.5	1480
2WSF		3700	000	284.9	1480
3WSF		3700	000	427.4	1480
5PSA	П	3515	185	000.0	1480
10PSA		3330	370	000.0	1480
1WSF5PSA	Ш	3515	185	142.5	1480
1WSF10PSA		3330	370	142.5	1480

Figure 9. Methods of testing: (a) flow table device, (b) compressive strength device, (c) flexural strength device

6. **Results and discussion**

6.1. Flowability and bulk density

The flowability of the grouts was measured by examining the flow table for all mixtures. Figure 10 shows the flowability results of the grout mixtures. The fluidity of the control mix that contains only cement is 24.8 cm, and the W/B ratio is 0.4%, but this fluidity decreases or increases according to the type of additives. When entering the proportion of WSF 1%, 2%, and 3% by volume, a decrease in the flowability of the grout appears at 23.1, 21.3, and 20.5 cm compared to the control mix. As the percentage of fibers increases, the flowability of the grout decreases [52, 53]. The reason for this is that the WSF was randomly distributed in the grout mixtures and acted as a skeleton, thus hindering the flow of the grout [54]. On the other hand, when replacing the OPC with PSA in proportions of 5% and 10% by weight, a decrease in the flow of the grouts is noticed. This is due to the surface area of PSA of 1517 m2/kg compared to OPC of 339 m2/kg. The greater the surface area, the greater its ability to absorb water, and thus the

decrease in the flow of the grouts [45, 55]. When WSF and PSA are inserted together with cement, the flowability of the grouts is significantly reduced compared to the control mix and for the same reasons mentioned. The bulk density results of the grouts, it is shown in Figure 11. Through the

results, it is clear that the bulk density increases with age, due to the continuation of the process of hydration of cement with water to complete the required reaction. When adding WSF with cement leads to an increase in density compared to the control mix because the density of WSF is higher than cement. But when PSA is introduced with cement, it leads to a decrease in density compared to the control mix because the density of PSA is less than cement. When adding WSF with PSA combined with cement, the density increases by a small amount compared to the control mix because PSA contributes to a decrease in density, unlike WSF.

6.2. Compressive strength

The compressive strength properties of sustainable grout cubes of dimensions $50 \times 50 \times 50$ mm3 and for ages 7 to 28 days were investigated, with three samples for each age. Figure 12 shows the compressive strength results of the control mix as well as mixtures containing different proportions of WSF and PSA. From the results shown below, it is evident that the compressive strength increases with age because the cement needs time to complete the hydration process and thus gives the required strength [56]. The results also showed that the compressive strength of 1WSF, 2WSF, and 3WSF mixtures is higher than the control mix for all ages. The reason is that when adding WSF to the cement will reduce the cracks that occur in the grout by binding the fibers to the cement paste [57].

It also binds the cement components through the pozzolanic reaction at an early age, producing large quantities of calcium silicate hydrate (C-S-H) gel, which is responsible for enhancing strength at an early age. Then, it binds the cement components, condensing the microstructure until the grout reaches its maximum resistance at the age of 28 days, thereby forming a high-performance structure that can withstand heavy loads. The increase in compressive strength at 7 days for mixtures 1WSF, 2WSF, and 3WSF was 20.89%, 12.29%, and 5.29%, respectively, while it was 8.86%, 6.22%, and 3% at 28 days. On the other hand, the compressive strength gradually decreases for mixtures 5PSA and 10PSA compared to the control mix. As the PSA affects the cement hydration process, its negative effect is reflected in the strength [58]. The reduction percentage at 7 days for the mixtures 5PSA and 1 PSA was 2.60% and 7.48%, respectively, while at 28 days it was 18.43% and 25.19%. But when 1% of WSF was introduced with PSA represented by mixtures 1WSF5PSA and 1WSF10PSA, a clear improvement in compressive strength is observed compared to mixtures that contain only PSA, but the strength remains less than the control mix.

6.3. Flexural Strength

Figure 13 shows the results of the flexural strength of the control mix and mixtures containing different percentages of PSA and WSF. These results were obtained through grouting prisms with dimensions (160 x 40 x 40 mm) for ages 7 and 28 days, with three samples for each age. From the results, it is clear that the flexural strength increases with an increase in the percentage of WSF for the mixtures 1WSF, 2WSF, and 3WSF compared to the control mixture because the WSF works to restrict the cracks that occur in the grout materials [59, 60].

Figure 10. Flow table test for different grout mixtures.

Figure 11. The bulk density results of various grout mixtures at 7 and 28 days of curing.

Figure 12. The compressive strength results of various grout mixtures at 7 and 28 days of curing

Figure 13. The Flexural strength results of various grout mixtures at 7 and 28 days of curing

Figure 14. The broken prism specimens after the test

The increase in flexural strength at 7 days was 37.46%, 21.89%, and 98.09%, respectively, while at 28 days it was 12.50%, 23.96%, and 44.96%, respectively. Furthermore, the flexural strength gradually decreases for the 5PSA and 10PSA mixtures that contain paper ash as a partial replacement for cement compared to the control mixture [61]. The percentage of decline at 7 days was 11.75% and 35.56%, while at 28 days it was 25.52% and 33.16%. When 1% of WSF was added with 5% of PSA, an increase in flexural strength of 10.16% and 2.95% was observed at the ages of 7 and 28 days, respectively, compared to the control mixture. However, when 1% of WSF was introduced with 10% of PSA, a decrease in flexural strength was noticed compared to the control mixture. Figure 14 shows the broken prisms of the grout mixtures after the test

7. Conclusions

This research attempts to prove the feasibility of preserving the environment by using recycling industrial waste and waste extracted from car tires as effective supplementary cementitious materials, that can be used in grouting for various civil engineering applications. Extensive laboratory tests were conducted, including mechanical and functional tests such as flow table, compressive strength, and flexural strength, to evaluate the characteristics and behavior of modified grout specimens with waste materials compared with conventional grout. The significant results obtained from conducting these tests are presented in the following points:

- Introducing WSF or PSA with cement leads to a decrease in the fluidity of the grouts. The mixture containing 3 % WSF, 10 % PSA, and 1 WSF and 10 PSA shows a decrease in flowability compared to the control mixture.
- Adding WSF with cement improves the bulk density compared to the reference mixture, unlike the addition of PSA which reduces it due to the uniform distribution of WSF within the mixture's texture of cementitious grout material. Combining WSF and PSAs with cement results in a slight increase in bulk density.

Partially replacing cement with WSF raises the compressive strength at all replacement ratios. For example, the mixture with 1% WSF showed a rise of 20.89% at 7 days and 8.86% at 28 days compared to the control mixture Mixture.

This is due to WSF's superior ability to bind the cement paste fibers and increase the hydration compounds, as well as the formation of CSH gel, which is responsible for increasing resistance. Replacing cement with PSA results in a decrease in compressive strength. However, combining WSF with PSA leads to an improvement in compressive strength, although it still falls short of the reference mixture's values.

- Flexural strength results showed similar compressive strength behavior for mixtures containing WSF. Mixtures with 1%, 2%, and 3% WSF replacement recorded increases of 37.5%, 21.9%, and 98.1% at 7 days of curing, and 12.5% a, 23.7%, %, and 44.9% at the age of 28 days, respectively. While mixtures containing PSA recorded a clear decrease in flexural strength. As for the mixture of PSA and WSF, the mixture containing 1% WSF with 5% PSA witnessed an increase of 10.2% and 2.9% at the age of 7 days and 8%, respectively, compared to the reference mixture.
- Mixing WSF and PSA with cement contributes to a slight improvement in the mechanical properties of cement grout.
- Based on all the tests in this study, it was found that the mixture containing 3% WSF and the mixture containing 1% WSF with 5% PSA are the optimal proportions for replacement, as they gave a clear improvement in workability, apparent bulk density, resistance to compression and flexural, and thus formed a strong cementitious grout skeleton that could withstand applied loads and prevent crack formation.
- It is recommended to use recycled industrial waste (such as waste steel fiber) in construction to improve its performance, reduce the demand for conventional resources, and reduce its harmful effects on the environment.

Authors' contribution

All authors contributed equally to the preparation of this article.

Declaration of competing interest

The authors declare no conflicts of interest.

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Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

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