

Original Research

Sustainable Concrete with Zero Carbon FootprintAhmed Alawais ^{*}, Roger P WestDepartment of Civil Engineering, Trinity College, University of Dublin, Ireland; E-Mails:
alawaisa@tcd.ie; rwest@tcd.ie^{*} **Correspondence:** Ahmed Alawais; E-Mail: alawaisa@tcd.ie**Academic Editor:** Mostafa Seifan**Special Issue:** [New Trends on Construction Technologies and Sustainable Building Materials](#)*Recent Progress in Materials*

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Received: April 15, 2024**Accepted:** August 04, 2024**Published:** August 09, 2024**Abstract**

This paper describes a method to use solely recycled and by-product materials as constituents to form concrete that can be used in buildings structural applications. As concrete is one of the most important materials in human civilization, where it is used widely in construction, cement and aggregate the main components of concrete cause an emission of large amounts of carbon dioxide, which is the main cause of global warming. The production of one tonne of cement, for example, causes the emission of about 800 kg of this CO₂. The growing demand for concrete constitutes a threat to the environment and its resources into the future. According to a market study by The Freedonia Group, in 2019 the world demand for cement was 5.1 billion tonnes which means that more than 2.5 billion tonnes of water and more than 11 billion tonnes of aggregates, both of which are scarce resources, will also be consumed. The goal of this Paper is to describe a 100% substitution of concretes normal constituents to form a sustainable concrete with zero carbon footprint and without compromising concrete mechanical properties. This will demand a pre-treatment of the recycled and by-products components to compensate for the natural strength loss due to their inclusion. Therefore, an innovative novel treatment method is selected for recycled concrete aggregates and chipped rubber to be separately treated and tested to mitigate the loss of strength in proposing a novel recycled activator for GGBS and silica fume. Then these waste recyclable materials are



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combined in a concrete mix that is 100% recycled and, therefore, significantly more sustainable.

Keywords

Sustainability; GGBS; concrete; cement; crumb rubber; silica fume

1. Introduction

Concrete is a key element in the construction and building industry and is the second most consumed material on Earth after water [1]. Recently, the global focus has shifted towards sustainability, aiming to reduce the carbon footprint and enhance the relationship between buildings and the environment. This is encapsulated in the concept of green buildings with zero carbon emissions and environmentally friendly cities that minimize pollution. The construction industry is a significant energy consumer, responsible for approximately one-third of total energy emissions and large quantities of carbon dioxide (CO₂) release. These emissions include embodied CO₂ from the entire building life cycle, from material production and construction to operational life and eventual disposal [2, 3]. However, incorporating recycled and by-product materials into concrete can compromise its strength and durability, necessitating innovative pre-treatment methods to mitigate these effects. The suggested pre-treatments include:

1.1 Impact of Pre-Treatment Methods on Concrete Properties

The usage of recycled and by-product materials in concrete can influence its properties significantly. Effective pre-treatment methods can enhance the quality and performance of concrete by addressing inherent weaknesses in these materials. The following pre-treatments are suggested:

- **Surface Treating Chipped Rubber (CR):** Treating chipped rubber from waste tires with ultraviolet light and sulfuric acid (H₂SO₄) enhances the stiffness and smoothness of the aggregates. This treatment improves the bonding between the rubber aggregates and cement paste, resulting in better mechanical properties and durability of the concrete.
- **Pre-Treating Recycled Concrete Aggregate (RCA):** Segregating, washing, drying, and surface treating RCA with micro silica fume (SF) reduces its porosity and enhances its strength. This process also improves the workability and compressive strength of the concrete, making it a viable replacement for natural aggregates.
- **Chemical Activation of Ground Granulated Blast Furnace Slag (GGBS):** Adding Lithium Chloride (LiCl) and Lithium Hydroxide (LiOH) as chemical activators accelerates the early-age strength development of GGBS. This enhancement allows for the use of GGBS in concrete without compromising initial strength, contributing to a more sustainable mix.
- **Using Waste Limestone Aggregate:** Incorporating 6.3 mm waste limestone aggregate as a coarse aggregate for particle packing optimizes the concrete mix's density and mechanical properties. This method reduces voids and enhances the overall strength and durability of the concrete.

- Utilizing Filtered Harvested Rainwater (HRW): Replacing normal tap water with filtered harvested rainwater in concrete production reduces the reliance on potable water resources and supports sustainability. The quality of HRW ensures it does not negatively impact the concrete's properties.

The research presented in this paper is crucial in advancing the sustainability agenda in the construction industry. By developing and implementing innovative pre-treatment methods for recycled and by-product materials, it is possible to produce concrete that maintains its strength and durability while significantly reducing its environmental impact. These pre-treatment techniques not only enhance the properties of recycled materials but also contribute to the overall goal of achieving zero-carbon footprint construction practices. This research underscores the importance of sustainable development and the potential for innovative solutions to drive progress in creating environmentally friendly buildings and cities.

2. Literature Review

2.1 Rubber in Concrete

About 1.4 billion tyres are sold each year and they fall into the waste category after the end of the tyre's life [4]. Regardless of the service life span of the tyres, it is estimated that every year almost one billion tyres end their service life of which more than 50% are dumped in landfills without any treatment [5]. Numbers are increasing because of the growing vehicle numbers and traffic worldwide [4, 6]. Studies have been conducted to develop techniques that combine concrete technology with chipped rubber [7]. Waste tyre rubber is added to concrete as a substitute for coarse and fine aggregate or sand. The studies revealed that the addition of chipped rubber to concrete could improve the elastic behaviour and sound insulation but reduced the compressive strength [6, 8, 9]. Nevertheless, suggestions were made on how to control the loss of strength; it might be minimized by a prior surface treatment with sodium hydroxide (NaOH_2) applied to the rubber particles to be used in the concrete [9]. Waste tyres are classified based on size to be used in concrete into shredded, chipped, crumb and ground rubber as explained below [10-14]. Figure 1 displays crumb and chipped rubber.

- Chipped rubber to replace coarse aggregate in the range 13-75 mm
- Crumb rubber that replaces sand in the range 0.425-4.75 mm
- Ground rubber that may be used as a fine filler in the range 0.075-0.475 mm

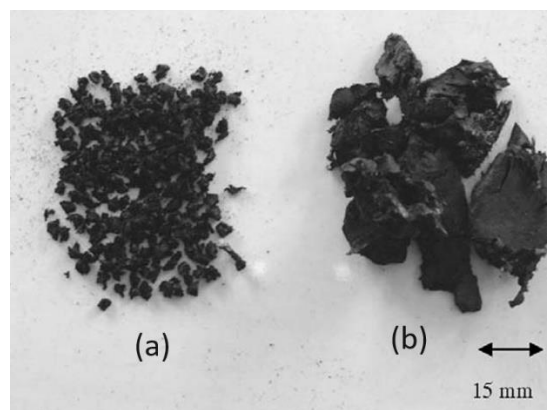


Figure 1 (a) crumb rubber, (b) chipped rubber particles [15].

2.1.1 Rubber Concrete Compressive Strength

Using recycled rubber in concrete usually leads to a lower concrete compressive strength [16]. This reduction in strength is caused by the hydrophobic nature of the rubber material and the stiffness difference between rubber particles and cement paste. The hydrophobic nature leads to imperfect bonding between the chipped rubber aggregate and cement paste where interfacial cracks can easily be generated under external load [17-19]. Furthermore, Reda Taha et al. [12] in their microstructural investigation on rubberized concrete, concluded that the reduction of strength might be attributed to the behaviour of the tyre rubber particles as a weak flexible aggregate, instead of the reduction of bond between rubber particles and cement paste.

Table 1 and Table 2 demonstrate that crumb rubber generally has a better compressive strength more than chipped rubber aggregates. However, there are instances where chipped rubber aggregate particles exhibit higher strength compared to some fine rubber concrete results at various replacement levels. It is important to note that numerous variables can influence the relative strength of rubber concrete compared to the control mix, such as the treatment method, mixing procedure, concrete slump/flow, curing conditions, and particle size distribution effects [20].

Table 1 Effect of chipped rubber size and replacement level on the 28-day compressive strength of rubber concrete [19].

Rubber Size (mm)	Replacement Level (%)	w/c ratio	Control mix Strength (MPa)	Strength relative to the control mix (%)
4-10	10, 15, 20, 25	0.40	43.5	69, 46, 34.5, 26.4
4-11.2	5, 10, 15	0.45	55.0	85.8, 68.5, 51.8
<12.7	25, 50, 75, 100	0.50	31.9	61.4, 43.3, 31, 23.5
<13	20, 40, 60, 80, 100	0.49	9.4	41.5, 34, 23.4, 10.6, 5.3
<15	12.5, 25, 37.5, 50	0.45	30.8	20.6, 4, 2.6, 1.8
5-20	10, 20, 40, 60, 80, 100	0.35	61.7	74.4, 53, 41, 25.6, 23.2, 14.1
5-20	25, 50, 75	0.52	45.8	52.2, 45.6, 38
5-20	25, 50, 75, 100	0.57	26.5	60.4, 52.1, 25.3, 21.5
<38	25, 50, 75, 100	0.48	33.7	55.8, 36.2, 26.4, 19.9
10-40	5, 10, 15, 20, 25, 30	0.40	54.0	88, 81.5, 70.4, 62.4, 57, 50.9
10-50	5, 10, 20, 30, 40, 50	0.48	37.5	73.3, 56, 33.3, 16, 9.9, 6.7
4.75-25	10, 20, 30, 40, 50	0.49	35.0	71.4, 51.4, 34.3, 8.6, 14.3

Table 2 Average relative strengths of fine and chipped rubber concretes at various replacement levels [20].

Rubber type	Replacement levels of conventional aggregates with waste tyre rubber (%)					
	5	10	20	30	40	50
	Relative strength of rubber concrete at various replacement levels \pm S.D.					
Fine rubber	87.1 \pm 5.2	76 \pm 7	59.5 \pm 13.4	43.9 \pm 14.9	32.3 \pm 14.5	39.7 \pm 27.8
Coarse rubber	82.4 \pm 65	69.9 \pm 8.4	42.5 \pm 10.5	33.5 \pm 17.5	28.3 \pm 13.3	31 \pm 19.5

2.1.2 Rubber Surface Treatment with Sulfuric Acid

The treatment of rubber with sulfuric acid (H_2SO_4) produces a noticeable decrease in contact angle which was mainly ascribed to an increase in surface energy due to the formation of sulfonic acid moieties and C=O bonds, and the removal of zinc stearate [21]. Moreover, the rubber surface swelled and became brittle and, when flexed, microcracks were created. Also, a rubber surface layer modification was produced with a consequent decrease in tensile strength, though it was not tested in concrete. This treatment enhanced the strength of the rubber's adhesive properties. The optimum immersion time in the H_2SO_4 solution was less than 1 minute and the reaction time in air was not found to be critical; the subsequent neutralization, with ammonium hydroxide, of the high concentration of the H_2SO_4 (95%) was essential in producing adequate effectiveness of the treatment [21, 22].

2.2 Recycled Concrete Aggregates

The use of recycled and by-product materials requires a strong understanding of their characteristics, which can be problematic because the inconsistency of the material characteristics can be high. Thus, innovative material pre-treatment methods can help to minimize this variability to some extent [23]. Furthermore, it is difficult to overcome the barriers that prevent the wider use of recycled aggregates in construction [24], where barriers are demonstrated in Figure 2.



Figure 2 Main barriers that prevent a wider use of recycled aggregates in construction [24].

A sustainable solution for the vast amount of waste debris is to recycle it as new concrete aggregate. Through a process of crushing waste concrete to produce recycled concrete aggregates to add in a new concrete mix, one can achieve a more environment-friendly concrete [25]. By using

a deconstruction approach of a building, it is likely to retrieve a greater number of reusable materials [24, 26].

Waste concrete can be crushed into either fine or coarse aggregates. Recycled concrete aggregate (RCA) are composed of adhered mortar and original aggregates. Thus, in comparison with normal aggregates, recycled concrete aggregates have a higher water absorption ratio and a lower specific gravity. Also, the crushing values and the Los Angeles abrasion percentages are much higher [27]. Therefore, the properties of RCA must be enhanced for it to work effectively in load-bearing structures [28-30]. The mechanical characteristics of RCA largely depend on the type of recycled aggregate used. RCAs are expected to result in an increase in the water absorption of concrete, at a rate directly proportional to the RCA content [31-33].

This paper will suggest a pre-treatment method to mitigate the water absorption in recycled concrete aggregate, that will ease the use of extra water which will benefit the strength of concrete made from recycled concrete aggregates.

2.2.1 Recycled Concrete Aggregates Compressive Strength

Concrete's mechanical properties are adversely affected by using recycled and by-product materials as substances, especially compressive strength, and modulus of elasticity, yet the reduction in these properties may be countered to some degree by pre-treating these substances [23, 32]. Replacing 50% to 100% of NA with RA decreases the compressive strength by 5% to 25%. However, up to 30% of NA can be substituted with coarse RCA without any effects on concrete strength. Strength gain for RCA is lower than normal NA concrete (Figure 3) [34, 35].

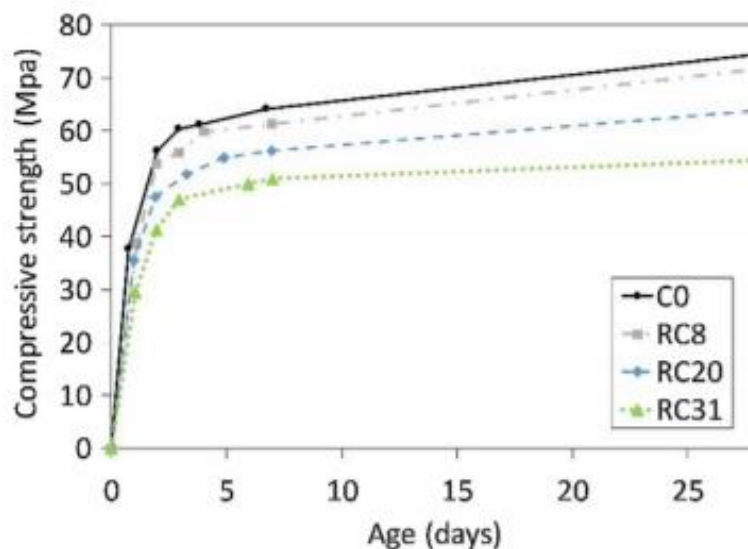


Figure 3 Compressive strength at early ages and at 28 days [35].

Rahal [25] illustrated that concrete with 100% replacement of recycled aggregates and a lower w/c ratio than normal concrete may have a higher compressive strength, yet it is less workable. However, if the w/c ratio is adjusted to maintain workability then it is expected that the compressive strength of the RCA will be lower. Scholars working with recycled materials in concrete often mention the process of mixing the recycled constituents to form concrete, and this process varies

slightly in different papers. This study will suggest a novel process that counters the loss of compressive strength and workability without the usage of a superplasticizer.

2.2.2 Recycled Concrete Aggregates: Pre-Treatment with Silica Fume

Bui et al. [36] proposed a treatment method for RCA as shown in Figure 4. Firstly, the recycled aggregates are soaked in sodium silicate (Na_2SiO_3) solution for pre-treating and then coated with various percentages of silica fume.

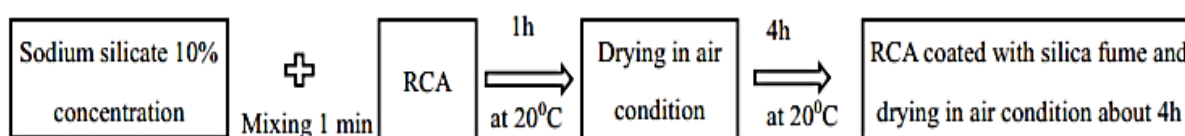


Figure 4 Pre-treating process for RCA with sodium silicate and silica fume [36].

Sodium silicate will penetrate the RCA structure to seal cracks and react with portlandite (calcium hydroxide $\text{Ca}(\text{OH})_2$ -CH crystals) of RCA to make a stronger C-S-H (calcium silicate hydrate $\text{CaO}\cdot\text{SiO}_2\cdot\text{H}_2\text{O}$) product in concrete as in the following reaction:



Sodium silicate is a commercial product that is used to activate pozzolans [37]. Sodium silicate is also used as an activator in geopolymer concrete [38]. Furthermore, sodium silicate can reduce the water absorption coefficient of RCA. Furthermore, silica fume can fill in pores and cracks of RCA and convert CH crystals into C-S-H gel [39-42].

This paper suggests a new process for coating RCA with silica fume, in includes draying the aggregates before coating and wating 28 day after coating for silica fume to set.

2.3 Ground Granulated Blast-Furnace Slag

Ground granulated blast furnace slag (GGBS) is a by-product of the iron industry, when heating the iron to about 1500°C a molten slag floats above the molten iron. Table 3 displays the chemical composition of GGBS.

Table 3 Chemical composition of GGBS and PC [42].

Chemical composition (wt.%)	GGBS	Cement
SiO_2	33.42	21.04
Al_2O_3	13.35	5.46
Fe_2O_3	0.21	2.98
CaO	41.16	63.56
MgO	7.76	2.52
SO_3	N/A	N/A
MnO	N/A	N/A
Others	4.10	4.44
Free-CaO	0.1	0.72

Cooling molten iron slag with water produces a glassy granulate, which is dried and then ground to the required size, to be known as ground granulated blast furnace slag [32, 43, 44]. It has a composition of 30% to 40% silicon dioxide (SiO₂) and approximately 40% CaO, which is less than the chemical composition of Portland cement (PC). GGBS is more environmentally friendly than Portland cement as its carbon footprint (circa 35 kg/m³) is considerably lower than PC (circa 800 kg/m³). The production of GGBS results in a release of up to 80% less CO₂ emissions than the emissions of CO₂ in PC [45, 46]. It has a better water impermeability and high resistance to corrosion caused by chloride ingress and sulphate attack. Although GGBS has a longer setting time, this can be an advantage because the concrete stays workable for a longer period and it reduces the risk of early thermal cracking [44, 47-50].

2.3.1 GGBS Effect on Compressive Strength

It takes time for the compounds in GGBS to react with water and obtain its hydroxyl ions whereby the hydration products of Portland cement break down the glassy slag parcels at an early age [51, 52]. Korde et al. [44] substituted up to 70% of Portland cement with GGBS and enhanced early age strength using a novel chloride free GGBS activator, a commercial accelerating admixture and by controlling the temperature. Table 4 displays the mix proportions. The 70% GGBS compressive strength results are displayed in Figure 5.

Table 4 GGBS mix properties [44].

Mix Proportions (per m ³)		Cementitious Materials Proportions		
Material	Mass (kg)	% GGBS	Cement (kg)	GGBS (kg)
Fine Aggregate	1758	0	586	0
Cementitious Materials	586	30	410.2	175.8
Water (w/c = 0.5)	293	50	293	293
Admixture (1% binder)	0 or 5.86	70	175.8	410.2

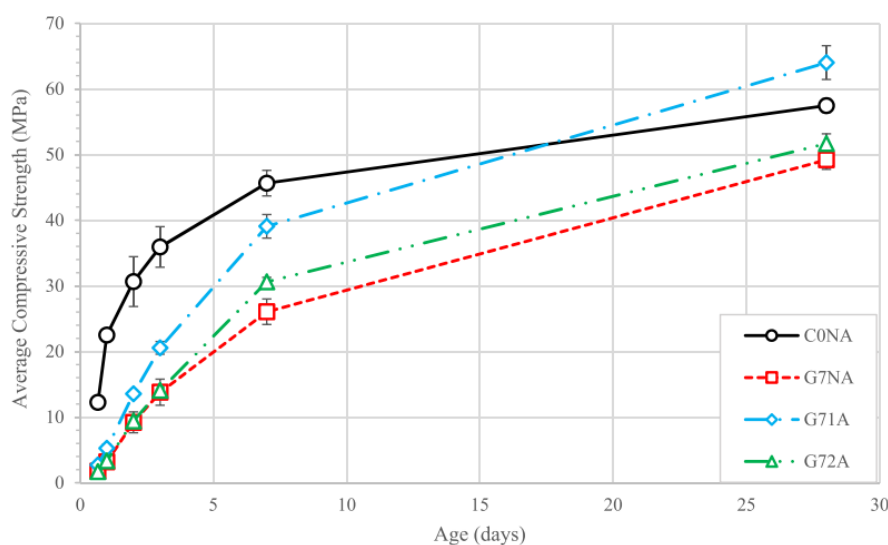


Figure 5 Relationship between CEM I 42.5 R and GGBS compressive strengths and age with and without admixtures for 70% slag at ambient temperature 0 = PC, 7 = 70% GGBS, NA = no admixture, 1A = new admixture, 2A = proprietary admixture [44].

With 70% GGBS and Admixture (1), the compressive strength development of the GGBS samples are considerably higher than without admixture or with (2). The main conclusion is that it is possible to achieve the equivalent early age strength of PC [44].

This work will replace 100% of the Portland cement and use 80% GGBS and 20% silica fume and will suggest a novel recycled Lithium-based activator to attain 28 days compressive strength as normal concrete without using temperature as an activator.

2.3.2 GGBS Alkaline Activators

GGBS is a slow reacting cementitious material, which can achieve a higher compressive strength when an alkaline activator is added to it. Kim et al. [53] classified alkaline activators into six groups: Caustic alkalis, non-silicate weak acid salts, Silicates, Aluminates, Aluminosilicates and non-silicate strong acid salts. The use of GGBS to replace Portland cement also increases the chloride binding capacity of the concrete. This is credited to their high alumina content. Chloride in concrete exists as free a chloride dissolved in the pore water, or as a bound chloride. The bound chlorides are either chemically bound to the tricalcium aluminate (C3A) and the calcium aluminoferrite (C4AF) or physically bound to the surface of the hydration products C-S-H gel [54-58].

This research will investigate using recycled lithium hydroxide and recycled lithium chloride as recycled alkaline activators for GGBS and silica fume.

2.4 Lithium

Recycling electronic waste to recover raw materials is a valuable approach to saving the environment. Electronic devices containing expensive metals such as lithium-ion batteries, which use lithium as the electro-active materials [59]. The demand for clean energy has significantly expanded the use of lithium-ion batteries (LIB) from applications such as small portable electronic devices all the way to electric vehicles [60, 61]. There is no shortage of lithium, however, there is shortage of highly pure lithium carbonate and lithium hydroxide [62, 63]. there are no papers in evidence which consider the use of Li as a GGBS/SF activator and this paper will consider this novelty.

2.4.1 Lithium Recycling

The process used to obtain lithium requires less energy, and less environmental impact compared to the extraction of it [64]. Most of the lithium resources for LIB are extracted from brine lakes which contain 66% of all lithium. Furthermore, there are many efforts to recycle metals from LIB, and recycled raw materials are regularly reapplied in new battery products (Figure 6) [59]. The recycling process in depth: the spent batteries are first discharged and then manually dismantled to recover the Al and Cu foils in metallic form and the separator, and then they are recycled after dismantling (Figure 7) [63].

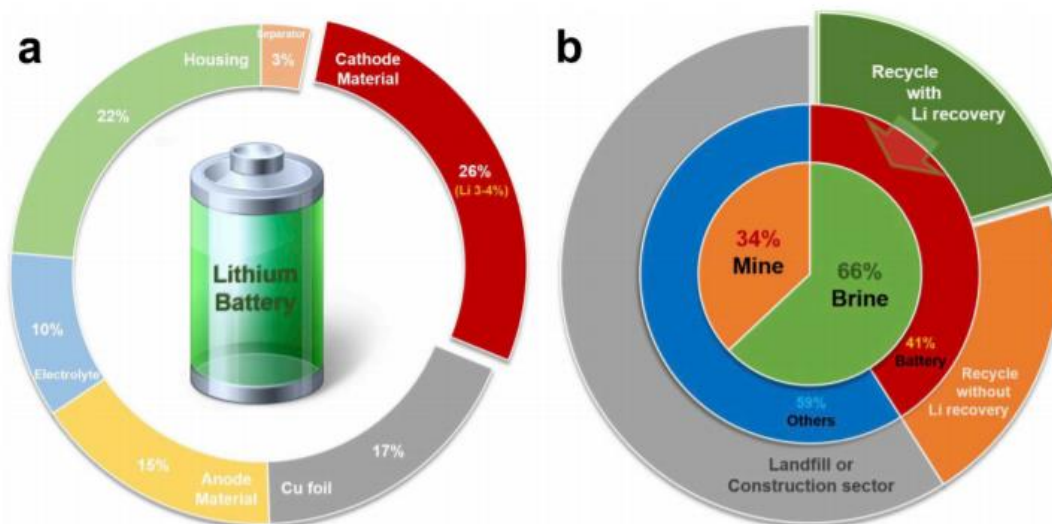


Figure 6 (a) Components of conventional lithium batteries (%). (b) Circulation of lithium resources [59].

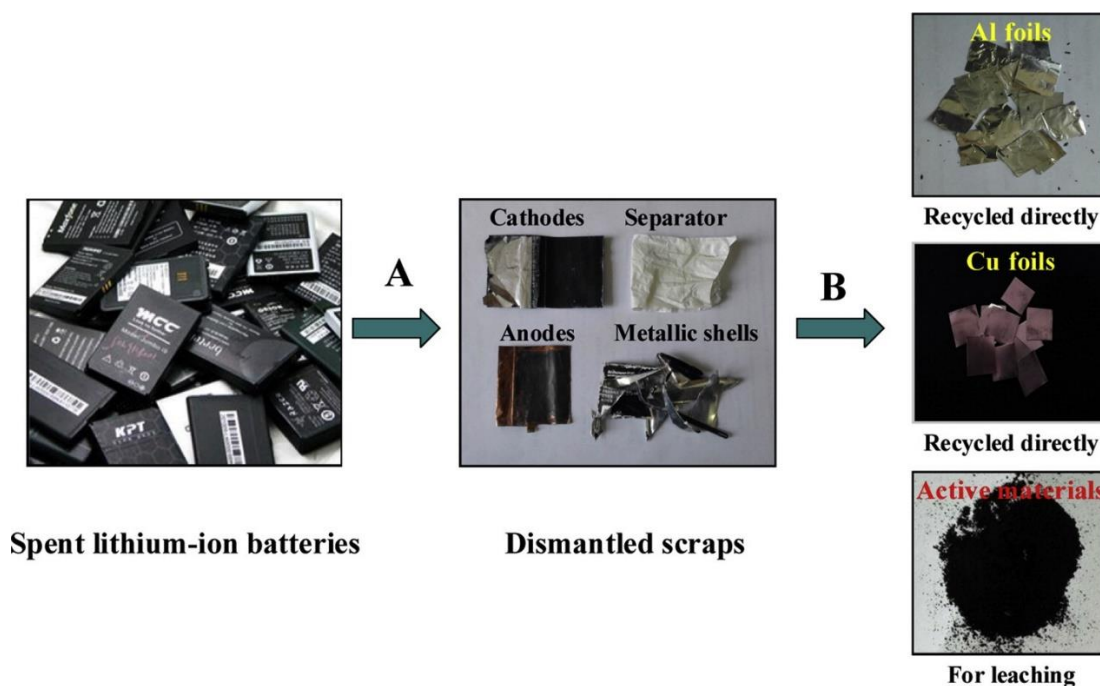


Figure 7 Simplified pre-treatment process of spent LIB based on citric acid/hydrogen peroxide oxidative leaching of Co and Li: (a) manual dismantling; (a) peeling off Al/Cu foils and recycling of Al and Cu [63].

2.5 Rainwater Harvesting

With increasing surface runoff flooding due to the vast urbanization contribution, rainwater harvesting is one of the best available methods for creating sustainable water cycles in urban developments [65]. Furthermore, Martin et al. [66] states that rainwater harvesting systems have two main elements:

- A catchment area to collect the rainfall.
- A large vessel that holds the harvested rainwater until it is called upon.

According to Chong et al., as cited by Leong et al. [67], a successful rainwater harvesting system involves gathering data on the physical, chemical, and microbiological characteristics of rainwater in order to minimise health risks from rainwater reuse and to design an effective water treatment system. Moreover, Leong et al. [67] concluded that regular cleaning of roofs and gutters is necessary for rainwater harvesting systems to diminish health risks from rainwater reuse. O'Hogain et al. [68] conducted a pilot study on rainwater harvesting in Ireland, where all results were obtained without any use of first flush devices or any form of disinfection. They illustrated that the physicochemical results complied with the drinking water regulations over the sampling period, except for ammonia. Table 5 shows results for the harvested rainwater compared with the S.I.278 [69] (European Communities Drinking Water Regulations) and S.I.155 [70] (European Communities Quality of Bathing Water Regulations).

Table 5 Overall physicochemical results for the harvested rainwater based on 14 monthly samples taken between January 2006 and January 2007 [71].

Parameter	Units	Mean	Min	Max	SD	Median	Drinking water guideline	Bathing water guideline
Chloride	mg/L	3.83	<0.01	27.83	7.37	1.49	250 mg/L	
Nitrate as NO ₃	mg/L	1.23	<0.01	2.84	0.92	1.20	50 mg/L	
Nitrite as NO ₂	mg/L	0.04	<0.01	0.20	0.06	0.03	0.50 mg/L	
Sulphate	mg/L	3.35	<0.01	37.40	10.26	0.30	250 mg/L	
Ammonia as NH ₃	mg/L	1.35	0.11	7.16	1.91	0.56	0.28 mg/L	
pH	pH units	7.07	6.67	7.83	0.31	6.98	6.5-9.5	6.0-9.0
TDS	mg/L	59.15	15.00	174.00	48.75	49.00		
TSS	mg/L	5.23	2.00	22.00	5.82	3.00		
Turbidity	NTU	0.63	<0.01	2.10	0.75	0.40	NAC and ATC	
Sodium	mg/L	2.62	<0.01	16.40	4.33	1.50	200 mg/L	
Calcium	mg/L	5.43	<0.01	46.80	12.89	1.40	None	
Lead, total	µg/L	3.28	<0.01	15.46	4.66	2.21	10 µg/L ^a	
Iron total	µg/L	61.50	<0.01	271.12	66.98	57.75	200 µg/L	
Cadmium, total	µg/L	<0.01	<0.01	<0.01	<0.01	<0.01	5.0 µg/L	

If the location of the rainwater harvesting system has a considerable number of birds, fecal materials from birds are usually present on the roof. That could probably be solved by increasing the size of the first-flush device. Alternatively, other measures, such as chlorination or UV radiation, should be applied if the harvested rainwater is to be used as potable water [72]. Note that Chloride levels are compatible with I.S.-EN-206 [73] (chloride content is shown in Table 6).

Table 6 Maximum chloride content of concrete [73].

Concrete use	Chloride content class ¹	Maximum Cl content by mass of cement ²
Not containing any steel reinforcement or other embedded metal with the exception of corrosion-resisting lifting devices	C1 1.0	1.0%
Containing steel reinforcement or other embedded metal	C1 0.2	0.2%
	C1 0.4	0.4%
Containing prestressing steel reinforcement	C1 0.1	0.1%
	C1 0.2	0.2%

This paper will investigate using harvested rainwater with a 100% replacement of normal tap water to in a concrete mix.

2.6 Particle Packing

Determination of concrete mixture constituents is one of the most important factors affecting the quality of concrete. It is possible to obtain a greater strength and quality in concrete with better packing of the different sized and shaped particles. In a concrete mix it is required to get an in-depth understanding of aggregates characteristics such as shape and size. The performance of concrete is significantly affected by the degree of packing of its aggregates. Thus, one needs an understanding of the concept of particle packing and its influence on concrete performance. The properties of aggregates have a significant role in determining the strength of concrete as the aggregate represents most of the concrete's volume. Aggregate is the main portion in forming the concrete. Appropriate aggregate blend characteristics are important to guarantee good concrete mixture performance [74]. Particle packing models are about selecting the proper sizes and proportions to fill voids. The smaller the particles the smaller the voids they contain, which are filled with smaller particles and so on. Kumar and Santhanam [75] demonstrated that research on particle packing involves selection of appropriate sizes and proportions of aggregate, to get a suitable combination for optimal packing and that is to:

- Understand how a specified combination of particles packed in a system works
- Develop mathematical models for calculating packing densities and porosities

The characterization of aggregate packing in concrete is not an easy task. Aggregate structure is a complex system, moreover packing aggregates can be affected by several properties such as the aggregate particle size distributions and shape [76].

Although some scholars approached aggregate particle packing as single sized and multiple sized spheres (Figure 8) like Mcgeary [77], Kolbuszewski and Frederick [78] and Mueller [79]. Yet, real aggregates are very much different from this model. Aggregates are irregular in shape and size [80].

¹ Chloride content classes are used to categorize the concentration of chloride ions in various substances.

² The maximum chloride (Cl) content by mass in cement is regulated to prevent corrosion of reinforcing steel in concrete structures.

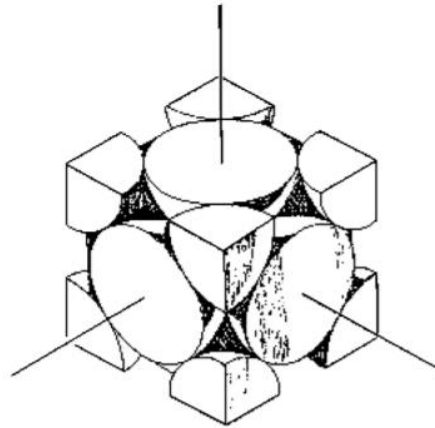


Figure 8 Maximum density arrangement of equal diameter spheres [80].

Dewar [81] simplified and explained the basic concept of the theory of particle package. He indicated that when two particles of different size are mixed, the smaller particle will attempt to fill the voids between the larger particles (Figure 9). The main relevant properties are mean size, voids ratio and relative density [81-83]. To understand the behaviour of any mixture of two sized particles three basic concepts are introduced by Dewar [82]:

- Only three parameters are needed to characterise any particle component (mean particle size, relative density, and voids ratio)
- The mixture can be studied by generating a void ratio diagram.
- The void ratio of the mixture is influenced by the void ratio of the components, particle interference and mean size ratio.

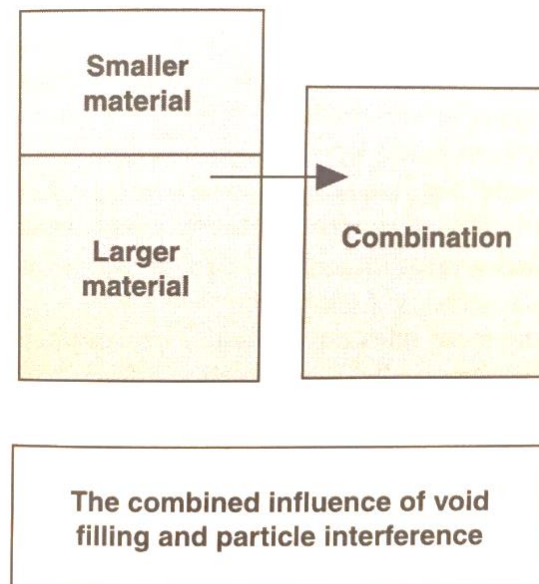


Figure 9 The basic principle of the Theory of Particle Mixtures [81].

This research will use the particle packing method to mitigate the strength loss when using recycled materials as concrete constituents.

3. Rationale

The study will examine the impact of using the recycled and by-products as concrete constituents, on the mechanical properties of concrete (strength, elastic modulus, workability, and density). This will generate a set of data that will be used to compare with other research and will be used as milestones in the evolution of this research. This study will examine the usage of the following in a concrete mix to be considered and used as a structural material:

- Chipped rubber to replace normal aggregates.
- 6.3 mm waste aggregate as an improvement for particle packing.
- Recycled concrete aggregates, coarse and fine, to replace normal aggregates.
- GGBS and silica fume as cementitious materials to 100% replace Portland cement.
- Harvested rainwater to replace normal tap water.

This study mitigated the negative effects on the concrete properties (strength, elastic modulus, workability, and density) of using the listed materials as follows:

- Pre-treatment of chipped rubber by submerging them into sulphuric acid to stiffen them and smooth their surfaces to get better workability and compressive strength.
- Using the 6.3 mm waste aggregate to make a better particle packed concrete, and then adding it with the same particle packing procedure to the pre-treated chipped rubber concrete for further improving the compressive strength of the rubberized concrete.
- Segregating recycled concrete aggregates based on strength, washing dust and quarry powder out of them, sieving them to the correct size, before coating them with silica fume with a suggested procedure to make a workable concrete that is as strong as normal concrete.
- Activating GGBS and silica fume with a new recycled material (lithium chloride and lithium hydroxide) to achieve a 28 compressive strength as a normal cement would achieve.
- Filtrating harvested rainwater before using it in a concrete mix.

This research will combine all these materials with the acquired knowledge to design a fully recycled by-product concrete mix. The materials will be combined in two mixes:

- a) Chipped rubber-based mix,
- b) Recycled concrete aggregate-based mix.

These two mixes are expected to be workable and have the required strength to be used in structural applications.

4. Experimental Program

For each mix, a total of 15 (300 mm × 150 mm) cylinders and 4 (100 mm × 100 mm) cubes were cast. The cubes were included to ensure independently that the two batches match in terms of their 28 days compressive strength. Five cylinders were tested respectively at 7, 28 and 56 days for strength and elastic modulus. Cubes were tested after 28 days for quality control.

5. Materials

5.1 Cement

This research project used Irish Cement bagged as a CEM II/A-L. The product has been specifically designed to reduce the carbon intensity of cement production by replacing about 14% of clinker

with ground limestone powder and to provide enhanced workability in block laying and plastering applications. The quality of all normal cement produced by Irish Cement is guaranteed to meet, in full, the requirements of Irish Standard I.S. EN 197-1 'Cement Part 1: Composition, Specifications and Conformity Criteria for Common Cements', and the product is independently certified by the National Standards Authority of Ireland and is CE marked.

5.2 GGBS

Ground Granulated Blast Furnace Slag (GGBS) used in this research was obtained from Ecocem GGBS which is produced by drying and grinding the GGBS at their milling plant in Dublin, manufactured to IS EN 15167-1. (<https://www.ecocem.ie/products/ecocem-ggbs/>)

5.3 Aggregates

Normal aggregates used in this thesis are produced at Roadstone Holding's limestone quarry, by means of blasting, crushing, and screening into the required sizes. Aggregates produced at their pit locations are extracted by excavator and screened/crushed into the required sizes: 10/14 mm, 14/20 mm and the waste aggregate 2/6.3 mm. Production in the roadstone quarry is managed under the ISO 9001 quality management scheme with all locations having either System 2+ or 4 levels of attestation of conformity as appropriate in accordance with IS EN 12620 Aggregates for Concrete and IS EN 13139 Aggregates for Mortars. Fine aggregates testing was incompatible with the ASTM International. Standard test method for relative density (specific gravity) and absorption of fine aggregate [84].

5.4 Recycled Aggregates

Recycled concrete aggregates were obtained by demolishing four concrete Kelly blocks that were cast for this project specifically to resemble a waste demolished building. Two of them had characteristic cube compressive strengths of 35 MPa compressive strength at 28 days and the other two were 50 MPa compressive strength at 28 days (Figure 10a). Kelly blocks were cored for compressive strength and density tests (Figure 10b). These blocks were crushed via the same mechanism used to crush normal aggregate (Figure 10c), that is to generate two batches different strengths of waste concrete in the form of RCA.



Figure 10 a) Kelly blocks, (b) Concrete cores (c) Aggregate crushing machine.

5.5 Silica Fume

Micro silica used in this study was acquired from Elkem Microsilica: 920 EN is a dry silica fume powder certified to IS EN 13263 'Silica fume for concrete'. Table 7 is the data sheet provided by the supplier.

Table 7 Chemical and physical properties of Elkem Microsilica® 920 EN are regularly tested in accordance with EN 13263.

Property	Unit	Value
SiO ₂	%	Minimum 85.0
SO ₃	%	Minimum 2.0
Cl	%	Minimum 0.3
Free CaO	%	Minimum 1.0
Free Si	%	Minimum 0.4
Loss on ignition	%	Minimum 4.0
Specific surface area	m ² /g	Minimum 15.0, Maximum 33.0
Pozzolanic activity index	%	Minimum 100

5.6 Chipped Rubber

Chipped rubber was obtained from a recycling car tyre centre, AES tyre recycling and remanufacturing facility, based in Co.Louth, which covers all aspects of the rubber tyre recycling process; from shredding, granulation and milling, to the production of saleable recycled products. Tyres are shredded down to 8-19 mm. (<https://www.aesirl.ie/business/tyre-recycling-remanufacturing/>)

5.7 Sulphuric Acid

Sulphuric acid used in this research was obtained from VWR chemicals

Formula: H₂SO₄

MW: 98.08 g/mol

Boiling Pt: 330°C (1013 hPa)

Melting Pt: 10.38°C

Density: 1.84 g/cm³ (20°C)

5.8 Lithium Hydroxide

Lithium hydroxide LiOH powder for this work was obtained from Alfa Aesar

Formula: LiOH·H₂O

MW: 41.96 g/mol

Boiling Pt: 920°C (1013 hPa)

Melting Pt: 462°C

Density: 1.51 g/cm³ (20°C)

5.9 Lithium Chloride

Lithium chloride LiCl powder for this work was obtained from Acros Organics

Formula: LiCl

MW: 42.39 g/mol

Boiling Pt: 1382°C (1013 hPa)

Melting Pt: 614°C

Density: 2.07 g/cm³ (20°C)

5.10 Concrete Testing Standards

To ensure the comprehensive testing of both fresh and hardened concrete, a range of standards and procedures are utilized. The determination of secant modulus of elasticity in compression is outlined in B.S.-EN-12390-13 Testing hardened concrete. Determination of secant modulus of elasticity in compression [85]. For testing fresh concrete, the sampling and common apparatus are specified in I.S.-EN-12350-1 Testing fresh concrete, Part 1: Sampling and common apparatus [86], while the slump test is detailed in I.S.-EN-12350-2 Testing fresh concrete. Part 2: Slump-test [87]. Additionally, the degree of compactability is measured according to I.S.-EN-12350-4 Testing fresh concrete. Part 4: Degree of compactability [88], and the density in fresh concrete is determined by I.S.-EN-12350-6 Testing fresh concrete. Part 6: Density [89]

For hardened concrete, standards include I.S.-EN-12390-1 Testing hardened concrete. Part 1: Shape, dimensions, and other requirements for specimens and moulds [90]. For the shape, dimensions, and other requirements for specimens and molds, and I.S.-EN-12390-2 Testing hardened concrete. Part 2: Making and curing specimens for strength tests [91] which covers making and curing specimens for strength tests.

The compressive strength of test specimens is addressed in I.S.-EN-12390-3 Testing hardened concrete. Part 3: Compressive strength of test specimens [92]. Furthermore, the tensile splitting strength of test specimens is specified in I.S.-EN-12390-6 Testing hardened concrete. Part 6: Tensile splitting strength of test specimens [93], and the density of hardened concrete is detailed in I.S.-EN-12390-7 Testing hardened concrete. Part 7: Density of hardened concrete [94].

6. Recycled Materials Pre-Treatments

6.1 Chipped Rubber Sulphuric Acid Treatment

Treating chipped rubber with sulfuric acid is a method used to enhance its properties for use in rubberized concrete mixes. This process involves several steps to ensure the rubber is adequately prepared to improve the performance of the concrete. The following outlines the step-by-step procedure for treating chipped rubber and preparing it for use.

Procedure:

- Soak the Rubber: Place the chipped rubber in a 95% concentrated sulfuric acid solution for 1 hour, stirring it during this time to ensure thorough exposure.
- Dry the Rubber: After soaking, let the chipped rubber dry in plastic containers for 24 hours to remove excess acid.

- Rinse the Rubber: Rinse the chipped rubber with water until there is no sulfuric acid smell. While water pH was not tested in this process, it is recommended to test the pH in a factory setting to ensure all acid is removed.
- Final Drying: Allow the chipped rubber to dry for one week before using it in rubberized concrete mixes to ensure it is completely dry and ready for use.
- Testing: Prepare three random batches of untreated chipped rubber to test their California Bearing Ratio (CBR) value (Figure 11) for baseline comparisons.



Figure 11 California bearing ratio (CBR) testing apparatus.

The sulfuric acid treatment process for chipped rubber involves soaking, drying, rinsing, and final drying to enhance the rubber's properties for use in concrete mixes. Proper testing and quality control measures, such as pH testing in a factory setting, are recommended to ensure the effectiveness of this treatment. By following these steps, treated chipped rubber can significantly improve the performance of rubberized concrete.

Furthermore, Figure 12 demonstrates the CBR results for three random H_2SO_4 treated chipped rubber samples. The plunger was set to move 1 mm every minute for 20 mm and a plot of force against displacement was produced. Sulfuric acid treatment has a more than 600% increase in chipped rubber stiffness.

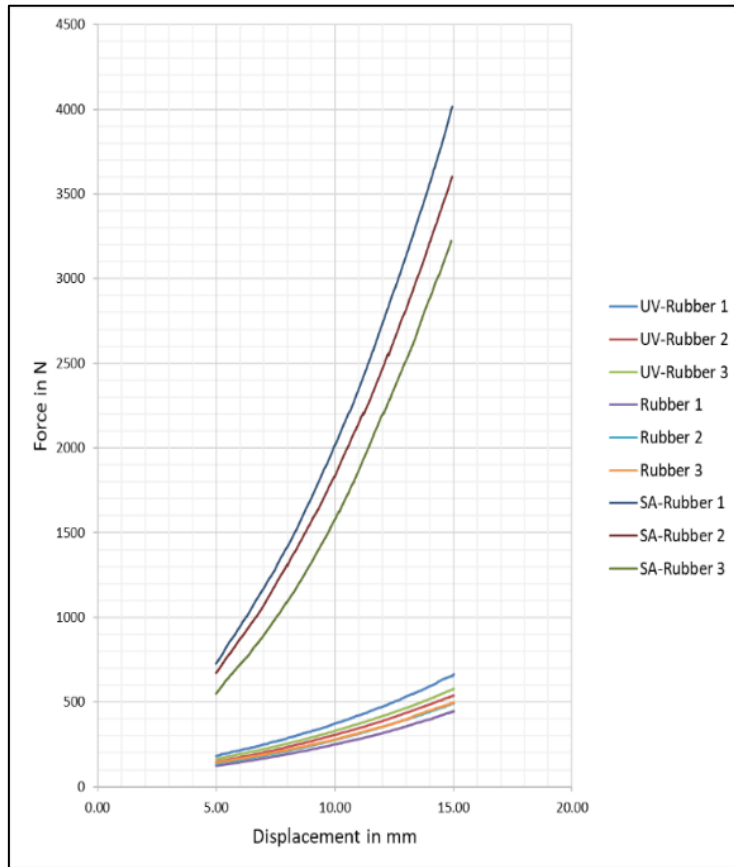


Figure 12 CBR results on treated and untreated chipped rubber with sulfuric acid, SA: sulfuric acid treatment, UA: ultra-violate light treatment.

6.2 Recycled Concrete Aggregates (Coarse)

Illustrated in Figure 13 is the recommended pre-treatment method by this paper, when using silica fume as a coating for recycled concrete aggregates. At first the recycled concrete aggregates are segregated based on their strength. Next, the aggregates are washed and cleaned from dust. Following, the aggregates are sieved to the required sizes then dried out to the lowest moisture content.

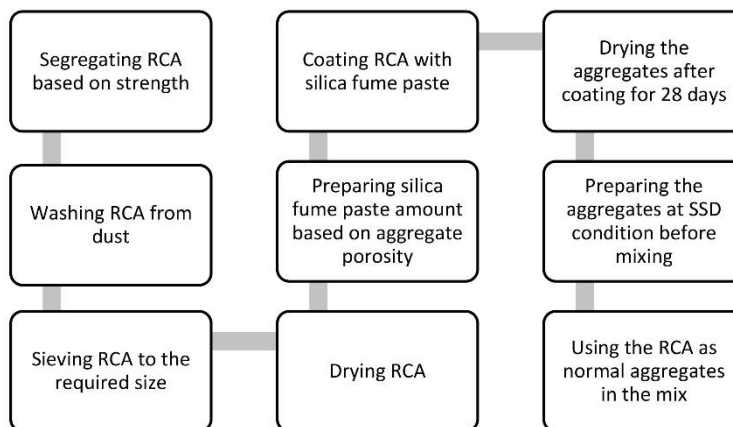


Figure 13 Recommended pre-treatment method.

Next, silica fume is prepared separately and mixed with the aggregates (Figure 14). After the aggregates are left for silica fume coating to set for 28 days. Recycled concrete treated aggregates are then ready to be prepared at SSD condition and to be used as aggregates in a concrete mix. Figure 15 displays RCA before treatment and 28 days after treatment. All of the surface irregularity and pores are coated with silica fume.



Figure 14 The steps of coating recycled aggregates with silica fume (1) dried aggregates, (2) preparing SF, (3, 4, 5) coating RCA with SF, (6) leaving the coated RCA to dry, (7) final product.



Figure 15 before treatment and 28 days after treatment.

6.3 Recycled Concrete Aggregates (Fine)

Coating recycled sand with silica fume (SF) is a method used to enhance its properties for use in concrete mixes. The process involves gradually adding SF to the sand until it reaches a saturated surface dry (SSD) condition, following the ASTM C 128 standard [80]. The following steps outline the procedure for coating recycled sand with SF (Figure 16).

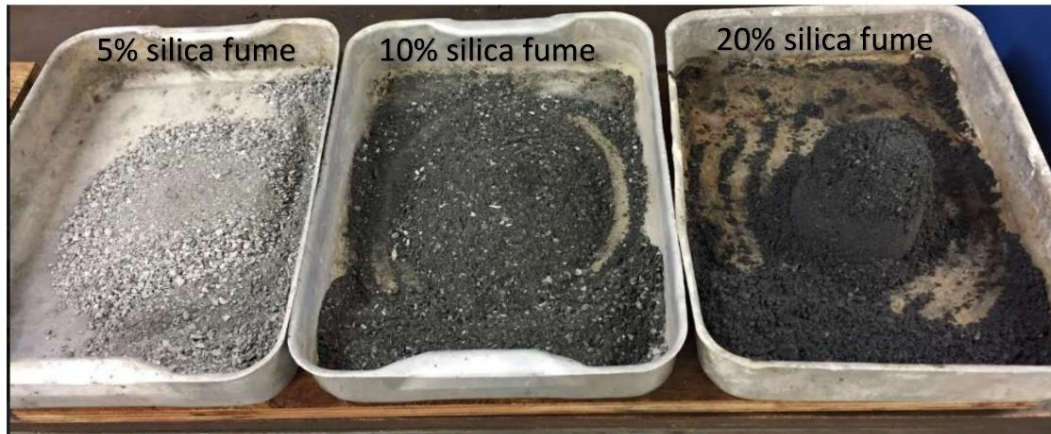


Figure 16 Recycled sand after coating with silica fume.

Procedure:

- **Dry the Sand:** Start by thoroughly drying the recycled sand to ensure it is free of moisture.
- **Add Silica Fume Gradually:** Begin adding silica fume to the dried sand gradually. The amount of SF added is based on trial and error until the sand reaches a saturated surface dry condition as defined by ASTM C 128.
- **Monitor SF Addition:**
 - a) **5% SF Addition:** Initially, add 5% SF to the recycled sand. At this level, there is minimal change in the condition of the sand, and it does not reach the SSD state.
 - b) **10% SF Addition:** Increase the SF to 10%. At this point, the sand replicates the saturated surface dry condition for fine aggregates, indicating an optimal level of SF.
 - c) **20% SF Addition:** Adding 20% SF results in oversaturation, failing the SSD test, as the sand becomes too wet.
- **Testing and Adjustment:** Continuously test the sand for SSD condition according to ASTM C 128 standard after each increment of SF addition until the desired SSD state is achieved.

By following these steps, recycled sand can be effectively coated with silica fume to reach the saturated surface dry condition, enhancing its suitability for use in concrete mixes. The trial-and-error method ensures the optimal amount of SF is determined, avoiding both insufficient and excessive addition. This process improves the properties of recycled sand, making it a valuable material in sustainable construction practices.

7. Sustainable Concrete Mix

Five concrete mixes were designed as displayed in Table 8. the mixes include a full replacement of the Portland cement, aggregate, sand, and water. The research investigates the combining the recycled, and by-product constituents with:

- chipped rubber (B1 and S1)
- recycled concrete aggregate (B2 and S2).

Table 8 Sustainable concrete mix, R: concrete normal mix, B1: base chipped rubber mix, S1: sustainable chipped rubber mix, B2: base recycled concrete aggregate mix, S2: sustainable recycled concrete aggregate mix, (a): 10% treatment of recycled sand, (b): 20% treatment of recycled sand, LC: lithium chloride, LH lithium hydroxide, SF: silica fume.

R	B1	S1	B2	S2	Constituents
700 kg/m ³	-	-	-	-	10 mm Normal aggregates
350 kg/m ³	-	-	-	-	20 mm Normal aggregates
-	125 kg/m ³	-	-	-	10 mm Chipped rubber
-	250 kg/m ³	-	-	-	20 mm Chipped rubber
-	-	75 kg/m ³	-	-	10 mm Chipped rubber treated with sulfuric acid
-	-	125 kg/m ³	-	-	20 mm Chipped rubber treated with sulfuric acid
-	-	-	700 kg/m ³	-	10 mm Recycled concrete aggregates
-	-	-	350 kg/m ³	-	20 mm Recycled concrete aggregates
-	-	-	-	700 kg/m ³	10 mm Recycled concrete aggregates (treated with SF)
--	-	-	-	350 kg/m ³	20 mm Recycled concrete aggregates (treated with SF)
-	-	500 kg/m ³	-	-	6.3 mm waste aggregates
700 kg/m ³	700 kg/m ³	--	700 kg/m ³	-	Normal Sand
-	-	700 kg/m ³ (b)	-	700 kg/m ³ (a)	Recycled Sand
-	-	80 kg/m ³	-	80 kg/m ³	Silica fume
-	-	320 kg/m ³	-	320 kg/m ³	GGBS
400 kg/m ³	400 kg/m ³	-	400 kg/m ³	-	Cement
-	-	40 kg/m ³	-	40 kg/m ³	LC
-	-	40 kg/m ³	-	40 kg/m ³	LH
200 L/m ³	200 L/m ³	-	200 L/m ³	-	Water
-	-	200 L/m ³	-	200 L/m ³	Rainwater

7.1 Fresh Sustainable Concrete Properties

Slump and density tests were conducted for all concrete mixes. Table 9 illustrates all density and slump readings for the five different mixes. The B1 mix showed significantly lower densities of 1585 kg/m³ and 1640 kg/m³ for batches 1 and 2, respectively. This reduction is expected due to the inclusion of chipped rubber aggregate, which has a lower density than traditional aggregates. Similarly, the S1 mix had densities of 2240 kg/m³ and 2265 kg/m³ for batches 1 and 2, respectively, still lower than the reference mix (R).

Table 9 Fresh sustainable concrete properties, R: concrete normal mix, B1: base chipped rubber mix, S1: sustainable chipped rubber mix, B2: base recycled concrete aggregate mix, S2: sustainable recycled concrete aggregate mix.

Mix	Density		Slump	
	Batch 1	Batch 2	Batch 1	Batch 2
R	2380 kg/m ³	2375 kg/m ³	145 mm	150 mm
B1	1585 kg/m ³	1640 kg/m ³	60 mm	65 mm
S1	2240 kg/m ³	2265 kg/m ³	65 mm	60 mm
B2	2263 kg/m ³	2282 kg/m ³	25 mm	25 mm
S2	2290 kg/m ³	2305 kg/m ³	40 mm	45 mm

In the S2 Mix, the replacement of Portland cement with Ground Granulated Blast-furnace Slag (GGBS) in caused about a 4% loss in density compared to the R mix, due to the lower specific gravity of GGBS. The densities for the S2 mix were recorded at 2290 kg/m³ and 2305 kg/m³ for batches 1 and 2.

There was and overall Decrease in Workability decreased by about 56% when all the constituents were added together. This is reflected in the lower slump values across all mixes with additional materials.

Observations:

- For the B1 mix, slump values were 60 mm and 65 mm for batches 1 and 2, respectively.
- The S1 mix showed slump values of 65 mm and 60 mm for batches 1 and 2.
- The B2 mix had the lowest slump values of 25 mm for both batches, indicating very low workability.
- The S2 mix had slump values of 40 mm and 45 mm, slightly higher than B2 but still lower compared to the R mix.

These results highlight the significant impact of incorporating different materials on the physical properties of concrete, particularly in terms of density and workability. The use of chipper rubber aggregate and GGBS notably influences these properties, providing essential insights for optimizing concrete mix designs.

7.2 Compressive Strength

The S1 chipped rubber-based mix exhibited a remarkable increase in compressive strength, rising by more than 600% at 28 days comparing it to B1. However, it still fell short of the baseline reference mix (R) in terms of strength. Despite not fully regaining the compressive strength due to the substitution of normal sand with recycled sand—which had a significant negative impact on compressive strength, the S1 mix still holds potential for structural applications based on its compressive strength performance.

On the other hand, the S2 recycled concrete mix showed a 23.5% increase in compressive strength at 28 days and a further 15% increase at 56 days. Impressively, by 56 days, the S2 mix not only matched but exceeded the reference mix's strength by about 8.5%, demonstrating its viability as an alternative to traditional concrete in terms of long-term strength.

Furthermore, several key insights can be drawn regarding the performance of different concrete mixes:

- **Material Substitution Impact:** The substitution of traditional materials with recycled and alternative materials significantly impacts the compressive strength of concrete. Mixes incorporating recycled materials, such as the B1 and B2 mixes, demonstrated notable variations in performance compared to the reference mix (R).
- **Early vs. Long-term Strength:** The initial strength at 7 days is markedly lower for mixes containing recycled materials. However, these mixes show significant strength gains over time, particularly evident in the S2 mix, which surpassed the reference mix's strength at 56 days.
- **Recycled Sand's Effect:** The inclusion of recycled sand had the most severe impact on the compressive strength, as seen in the S1 mix. This suggests that while recycled sand can be used, it may require further processing or combined with other materials to enhance its performance.
- **Chipped Rubber Aggregate:** The use of chipped rubber aggregate in the B1 mix resulted in much lower early strength but displayed a considerable increase by 56 days. This indicates potential for specific applications where long-term strength development is acceptable.
- **Structural Viability:** Despite some mixes not matching the reference mix's early strength, their significant gains over time suggest potential for structural applications. Particularly, the S2 mix's performance highlights its suitability for long-term structural use, where initial strength may be less critical.
- **Sustainability Considerations:** The successful use of recycled materials in concrete mixes, as demonstrated by the S1 and S2 mixes, underscores the potential for more sustainable construction practices. These mixes not only divert waste from landfills but also reduce the reliance on natural materials, contributing to environmental conservation.

Overall, these findings provide valuable insights for the development and optimization of concrete mixes that balance performance with sustainability, for more eco-friendly construction solutions (Figure 17).

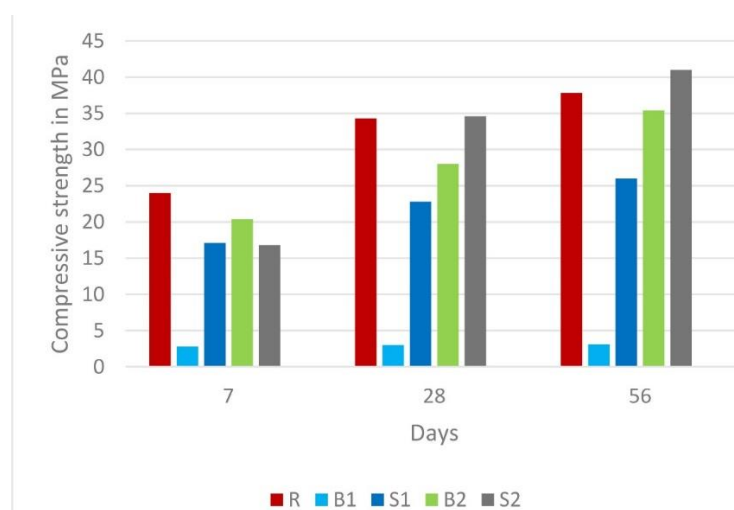


Figure 17 Sustainable concrete compressive strength, R: concrete normal mix, B1: base chipped rubber mix, S1: sustainable chipped rubber mix, B2: base recycled concrete aggregate mix, S2: sustainable recycled concrete aggregate mix.

7.3 Sustainable Concrete Elastic Modulus

Elastic modulus results are illustrated in Table 10. The 28-day result for the S1 chipped rubber sustainable mix showed an elastic modulus about 25% lower than the reference mix (R). This reduction is expected due to the inherently low modulus value of chipped rubber concrete. However, compared to the B1 mix, the elastic modulus of S1 increased by approximately 1060% at 28 days. On the other hand, the B2 sustainable concrete combined mix had an elastic modulus at 28 days that was about 13% lower than the reference mix. Despite this reduction, the use of a GGBS/silica fume binder in the S2 mix compensated for any loss in elastic modulus. Consequently, the elastic modulus for the S2 mix was about 15% higher than that of the B1 mix, attributed to the effective aggregate particle packing.

- Reference Mix (R): The reference mix maintains the highest elastic modulus values at all measured time points.
- The B1 mix shows very low elastic modulus values at all ages, with minimal improvement over time.
- S1 Mix: Although lower than the reference mix, the S1 mix demonstrates a significant improvement over the B1 mix, indicating the potential for applications where a balance between sustainability and performance is required.
- The B2 mix, while lower than the reference mix, shows substantial improvement and indicates that combining sustainable materials can yield competitive results.
- The S2 mix outperforms the B1 mix significantly and closely approaches the performance of the reference mix by 56 days, showcasing the benefits of using a GGBS/silica fume binder and effective aggregate particle packing.

Table 10 Sustainable concrete elastic modulus, R: concrete normal mix, B1: base chipped rubber mix, S1: sustainable chipped rubber mix, B2: base recycled concrete aggregate mix, S2: sustainable recycled concrete aggregate mix.

Mix	7 days	28 days	56 days
R	23.1 GPa	28.0 GPa	29.3 GPa
B1	1.2 GPa	1.8 GPa	2.0 GPa
S1	18.1 GPa	20.9 GPa	21.0 GPa
B2	20.2 GPa	24.8 GPa	25.1 GPa
S2	25.0 GPa	28.8 GPa	29 GPa

These observations highlight the diverse impacts of different recycled and by-product constituents on the elastic modulus of concrete, offering valuable insights for optimizing sustainable concrete formulations to achieve desired performance characteristics.

8. Conclusions

The development of a novel sustainable concrete mix using recycled and by-product materials was accomplished by examining, treating, and testing each constituent separately. This approach helped to identify and address the weaknesses of each material before combining them into a single mix. The utilization of particle packing of the aggregates further contributed to the success of this

innovative concrete mix. When replacing normal aggregates with 100% chipped rubber aggregates, it is essential to batch the aggregates by volume rather than weight. This is because chipped rubber aggregates have a lower density, resulting in a greater volume for a given weight compared to traditional aggregates. On the other hand, untreated rubber concrete mix tends to be less workable due to the characteristics of chipped rubber, such as lower surface bond and water repellence. Additionally, chipped rubber is lighter, which further reduces the workability of the concrete mix. Thus, sulfuric acid treatment significantly improves the stiffness of chipped rubber aggregates. This treatment eliminates the angularity of the chipped rubber, making their surfaces smoother and enhancing their performance in the concrete mix.

Introducing 6 mm limestone as a partial replacement for chipped rubber in the sulfuric acid-treated mix increases the strength of the concrete. However, it also decreases workability due to the larger surface area of the 6 mm aggregates, despite their higher density. Pre-treating recycled aggregates with silica fume (SF) improves the workability of RCA concrete. The treatment reduces the porosity of the recycled aggregates, as the micro silica particles fill the surface capillary pores, enhancing the overall performance of the concrete mix. Furthermore, drying the recycled concrete aggregates (RCA) before treating them with micro silica fume significantly enhances the compressive strength of the concrete. The better penetration of SF results in a more effective coating, which reduces water absorption on the RCA surface, thereby improving the concrete's strength.

Achieving this level of performance means that concrete can be produced by substituting 100% of its constituents with recycled, waste, and by-product materials without compromising its compressive strength and elastic modulus. As a result, this innovative concrete can be considered a zero-carbon footprint material, contributing to more sustainable construction practices.

Author Contributions

Ahmed Alawais: Conceptualization, Writing original draft, Formal analysis, Review and editing, Experimental program conduction. Roger West: Methodology, Experimental Program design, Result review, Review and editing.

Competing Interests

The authors have declared that no competing interests exist.

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