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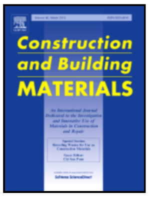

# Effects of pre-treatment using waste quarry dust on the adherence of recycled tyre rubber particles to cementitious paste in rubberised concrete

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## ABSTRACT

This research is focused on improving the mechanical response of rubberised concrete, with the objective of enhancing its weak Interfacial Transition Zone (ITZ). Pre-treatment of recycled tyre rubber particles by another material that is otherwise considered as waste, Waste Quarry Dust (WQD), diminished the hydrophobicity of rubber and subsequently eliminated excessive fluid repelling during mixing, key in reducing the gaps observed at the interface via SEM imaging. The pre-treatment consequently allowed for improved compressive strength; results indicate an increase of 282% and 276% in the 7 and 28-day compressive strength, respectively, for the selected rubberised concrete mixture.

## 1. Introduction

Research on the use of End-of-Life Tyre (EoLT) materials in concrete began in the early 1990s. The first studies on the inclusion of rubber particles in concrete [1,2] were published following the development of rubberised asphalt. Nowadays, while the world is shifting towards a circular economy [3], research [4–6] on rubberised cementitious materials continues, aiming to develop high added-value applications and concurrently assist with a critical environmental issue by fostering the re-use of a significant volume of tyre waste in the construction sector. Moreover, management of EoLTs is a major environmental concern worldwide; the European Union has outlawed the stockpiling of EoLTs through implementation of the EU Waste Legislation [7,8], due to their aesthetically unpleasing nature and imposed health threats [9–11]. In North America, the enforced “Scrap Tyre Regulations” vary by State; certain states have banned only whole tyres, while other states prohibit all tyres from landfills, and some states allow processed tyres into monofills only [12]. In Australia, data from national studies [13] on EoLTs reveal that 60–65% of all waste tyres generated, end up with little or no resource value recovered; they are either disposed to landfill, illegally dumped or stockpiled.

Recycling of tyre rubber in construction materials and particularly in concrete, the second most consumed material in the world after water [14], has been suggested [9,10,15] as an effective solution to the issue of EoLTs, since large volumes of waste tyre rubber can be exploited through this route. Inclusion of rubber particles in concrete and cementitious materials does not simply serve an environmental intent, but also conveys unique properties [16,17] to a material that is otherwise non-deformable and has low damping and energy absorption potential, thus enabling the development of innovative applications of high added-value and significant societal impact, such as forgiving surface transport infrastructure [18].

Among the products of tyre recycling is also steel wire, an exceptional source [19] of tensile reinforcement for concrete, if cleaned and sorted to specified lengths. Research [20,21] has shown that using recycled tyre steel as fibre reinforcement provides concrete of equal or better properties compared to ordinary fibre-reinforced concrete with manufactured steel fibres. While allowing efficient concrete micro crack controlling prior to larger crack formation, Recycled Tyre Steel Fibres (RTSF) (Fig. 1), increase concrete toughness [22].

RTSF are successfully [23] used in combination with tyre rubber particles in concrete, allowing the development of concrete and cementitious mortars with exceptional deformability and energy absorbing capacities [8,24]. While previous research on rubberised concrete [4,25,26] has identified the material’s strengths and promising behav-

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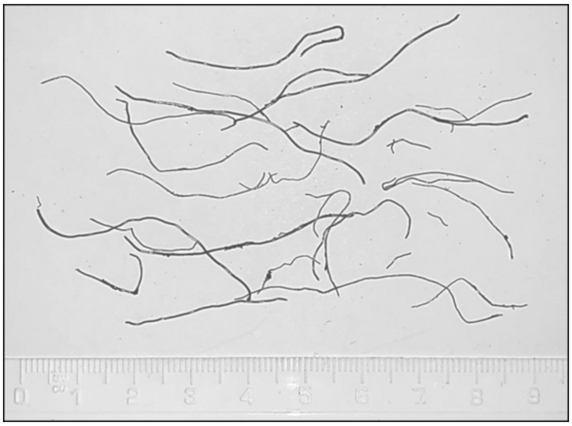

**Fig. 1.** Cleaned and sorted RTSF.

iour, certain aspects of its performance have not been addressed yet [18,27].

The major drawback of rubberised concrete is the significant reduction in compressive strength [28,29], with increasing rubber content. Although rather expected due to much higher Poisson's ratio of the rubber in comparison to the mineral aggregates being replaced, the reduction in strength is also attributed to poor adhesion of the rubber particles to the cement paste [30], also referred to as the weak Interfacial Transition Zone (ITZ). Lack of bonding at the ITZ generates gaps that are visible to the naked eye, while Scanning Electron Microscopy (SEM) images reveal higher porosity and limited formation of hydration products around the rubber particles [30].

The poor adhesion observed is attributed to the hydrophobicity [31] of rubber, which causes excessive fluid repelling during mixing, therefore the formation of air voids within the mixture, particularly at the rubber-cement paste interface [32,33]. This is justified by experimental findings of this study, further discussed in section 3.

To tackle the poor adhesion, several studies have examined rubber surface treatment using NaOH [34,35], pre-coating with mortar, cement paste [36] or silica fume [30], limestone powder pre-coating [37], UV radiation [38], partial oxidation [39] of rubber particles, using organic sulfur compounds [40] or silane coupling agents [41,42] as well as simply washing with water [30].

In terms of enhancing the hydrophilicity of rubber, surface treatment with NaOH solution was considered most promising [43], but in general, compressive strength increase was limited to 40%, therefore pre-treatments were considered unworthy of the cost and effort involved [30]. However, this research examines the potential of quarry dust, a material otherwise considered as waste, to tackle the rubber adhesion issue; aiming to provide a green, inexpensive solution to the weak ITZ of rubberised concrete. Waste Quarry Dust (WQD) accumulation is yet another rapidly growing environmental problem and health hazard [44] that could be controlled by WQD reuse in construction material. In addition to WQD use in brick making [45], research has been conducted on cement replacement with WQD [46,47], replacement of limestone fines [48] or using WQD as a supplementary cementitious material (SCM) [49] for sustainable concrete.

This study examines the use of WQD in pre-treatment of recycled tyre rubber particles, used as aggregate replacement in concrete. The significance of this research is entailed in the effectiveness of the novel treatment technique to minimise the hydrophobicity of rubber and therefore decrease fluid repelling during mixing, further reducing air void formation and improving rubber-cement paste adhesion. This facilitates an increase in the compressive strength of rubberised concrete, simply by the use of another waste material. The experimental "proof of concept" study is presented herein.

## 2. Materials and methods

### 2.1. Materials

All mixtures produced as part of this study included EN 197–1 [50] portland cement CEM I 52,5N supplied by Vassiliko Cement Works and "Tsirco-Microsilica'' or silica fume of 250–350 kg/m$^3$ bulk density, conforming to ASTM C1240 [51] and EN13263-1:2005 + A1:2009 [52] requirements for Silica Fume-Class 1. Crushed fine mineral aggregate from both diabase and limestone sources supplied by local quarries and coarse mineral aggregate from a local diabase quarry were used. The rubber particles used in this study were obtained by 3 different tyre recycling plants from Croatia, Cyprus and the U.K., having no specified properties. A representative sample of the rubber particles was tested following EN 1097–6 [53], to reveal an average specific gravity of 0.8. For both mineral and rubber aggregates, a fine to coarse aggregate ratio of 1.22 was kept. Commercially cleaned, sorted and evaluated RTSF of 2,560 ± 550 MPa [19] tensile strength and Young's Modulus of 200 GPa [19] supplied by Twincon Ltd. were used as fibre reinforcement. In addition, liquid polycarboxylic, polymer-based superplasticizer "Tsirco-Flo SCA 43H" of specified density of 1070 kg/m$^3$ ± 30 kg/m$^3$ and conforming to EN 934–2:2009 [54] was used. Mixtures NanoP and NanoS, as referred to in `Table 1`, included silica nanoparticles of 15–20 nm diameter, P-type and S-type, respectively, targeting an enhancement of the ITZ by the nanoparticle filling effect and not by pre-treatment as opposed to the first mixture variation. Both silicon oxide nanopowders were obtained from US Research Nanomaterials, Inc. The P-type ($SiO_2$, 99.5 + %, 15–20 nm, Porous) nanopowder used had a specific surface area (SSA) of 640 m$^2$/g, while the S-type ($SiO_2$, 99.5 + %, 15–20 nm, Spherical) nanopowder used had an SSA of 170–200 m$^2$/g. Fine quarry powder from a diabase source was used for pre-treating rubber particles, supplied by a local quarry; representative of the material that quarries consider as waste and typically discard.

### 2.2. Pre-treatment technique

Pre-treatment of rubber particles was achieved by surface coating with a paste, consisting of WQD and water. All rubber particles required for the mixture were weighed out separately by gradation

**Table 1**
Steel fibre reinforced rubberized concrete mixture constituents (1st set of mixtures).

| | Mixture IDs:<br>Control | NanoP | NanoS | Treated |
|---|---|---|---|---|
| Mixture Constituent | Quantity per m $^3$ of mixture | | | |
| Cement CEM I 52,5N | 400.0 kg | 400.0 kg | 400.0 kg | 400.0 kg |
| Silica Fume (Micro-silica) | 100.0 kg | 97.0 kg | 97.0 kg | 100.0 kg |
| P-type Nano-silica (15–20 nm) | – | 3 kg | – | – |
| S-type Nano-silica (15–20 nm) | – | – | 3 kg | – |
| Fine Crushed Aggregate (0–4 mm) | 310.5 kg | 310.5 kg | 310.5 kg | 310.5 kg |
| Coarse Crushed Aggregate (4–20 mm) | 378.0 kg | 378.0 kg | 378.0 kg | 378.0 kg |
| Fine Tyre Recycled Rubber Particles | 169.7 kg | 169.7 kg | 169.7 kg | 169.7 kg |
| Coarse Tyre Recycled Rubber Particles | 207.0 kg | 207.0 kg | 207.0 kg | 207.0 kg |
| Recycled Tyre Steel Fibres | 25.0 kg | 25.0 kg | 25.0 kg | 25.0 kg |
| Water | 280.0 kg | 280.0 kg | 280.0 kg | 280.0 kg |
| Super-plasticiser | 4.875 L | 4.875 L | 4.875 L | 4.875 L |
| Pre-treatment of rubber particles - Applied? | NO | NO | NO | YES |

size and then mixed together in a large bucket. Subsequently, the pre-treatment paste was prepared by mixing together equal parts of powder (WQD) and water, by mass. The dust and water were stirred thoroughly in a small bucket, and after reaching homogeneity, the slurry-like liquid was poured into the larger bucket, over the rubber particles. A few minutes of continuous stirring allowed the paste to coat the rubber particle surfaces thoroughly, before they were scooped out and laid on plastic sheathing to let the paste dry, as shown in Fig. 2. Fig. 3 shows an indicative sample of rubber particles covered by the pre-treatment paste a few minutes after treating (Fig. 3(a)), and 24 h after treating (Fig. 3(b)).

### 2.3. Steel fibre reinforced rubberized concrete

To investigate the effects of the rubber particle pre-treatment with WQD as well as the effects of replacing part of the microsilica included in the original mix with nanosilica, a Steel Fibre Reinforced Rubberized Concrete (SFRRC) mixture with 60% by volume replacement of aggregate by recycled tyre rubber particles was adopted. The original mix was optimized by the corresponding author for the development of SAFER road barriers [55] and was adjusted accordingly for this specific study.

Standard concrete cube specimens (15x15x15cm$^3$) were prepared, cured [56] and tested for Compressive Strength [57] following standard procedures. A 60-Litre capacity drum mixer was used to make the SFRRC mixtures. The recycled tyre rubber particles were added to the mixer first, together with the mineral aggregate. After mixing all aggregates for a few minutes, half of the mixture water was added and blending proceeded. The binder content was then introduced to the mix, followed by the rest of the water and superplasticiser. The RTSF were added last, before the final 3 minutes of mixing. The cube samples were consolidated using a vibrating table and left in their casting moulds for 48 h, then placed in a water tank at controlled temperature of 18–20 degrees Celsius, where the hardened cubes cured for 5 or 26 days, according to their day of testing. The cube specimens were all taken out of the water tank 48 h before their strength tests.

**Fig. 2.** Coated rubber particles on plastic sheathing.

#### 2.3.1. 1st set of mixtures

Initially, a set of 4 mixtures (Table 1) was investigated, aiming to compare 3 variations to a "control" mix, in which no rubber pre-treatment was applied and the binder content was limited to cement and microsilica. This 1st set of mixtures compares the effect of (1) pre-treatment of rubber particles to (2) including silica nanopowder in the ruberised concrete mix binder content, on the compressive strength of the material. Including silica nanopowder, although an expensive material and not readily available in many countries, is an established technique, proven to increase the compressive strength of conventional concrete. Thus the 1st set of mixtures aims to evaluate the increase in compressive strength achieved by pre-treatment of rubber particles, as opposed to an expensive technique. All 4 mixtures were of equal water to binder ratios (w/b) and had 60% by volume of their aggregate content replaced by equivalent size recycled tyre rubber particles. The first mixture variation (treated), included rubber particles that were pre-treated with WQD, and only cement and microsilica as binders, matching the control mixture's content. The second and third mixture variations included non-treated rubber particles, with the binder content modified compared to the control mix, in both cases by replacing 3% by mass of the microsilica content by nanosilica.

#### 2.3.2. 2nd set of mixtures

Experimental findings from the 1st set of mixtures indicated promising enhancement but high variability in the compressive strength of the mix that included rubber pre-treated with WQD, therefore a second set of mixtures followed. The 2nd set serves as direct comparison between using non treated rubber versus rubber pre-treated with WQD. The 2 mixtures of the 2nd set were prepared using identical material from the same sources and batches as the 1st set mixtures. Table 2 lists the constituents of the 2 mixtures (Non-treated and Treated), comprising the 2nd set. For the 2nd set of mixtures, instead of keeping equal w/ b ratios, the 2 mixtures were designed targeting identical flowability,

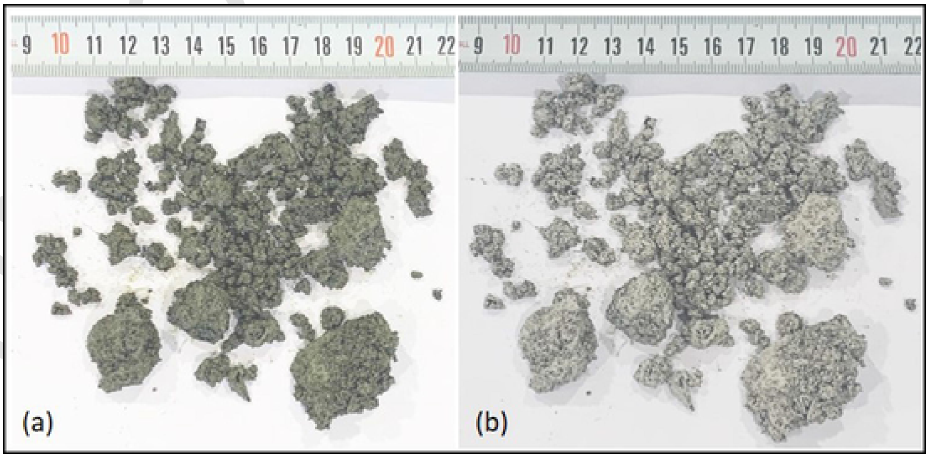

**Fig. 3.** Indicative sample of pre-treated rubber particles (a) a few minutes after pre-treatment and (b) 24 h after pre-treatment (scale provided in cm).

**Table 2**
Steel fibre reinforced rubberized concrete mixture constituents (2nd set of mixtures).

| | Mixture variation IDs: | |
|---|---|---|
| | (a) Non-Treated | (b) Treated |
| Mixture Constituent | Quantity per m $^3$ of mixture | |
| Cement CEM I 52,5N | 400.0 kg | 400.0 kg |
| Silica Fume (Micro-silica) | 100.0 kg | 100.0 kg |
| Fine Crushed Aggregate (0–4 mm) | 310.5 kg | 310.5 kg |
| Coarse Crushed Aggregate (4–20 mm) | 378.0 kg | 378.0 kg |
| Fine Tyre Recycled Rubber Particles | 169.7 kg | 169.7 kg |
| Coarse Tyre Recycled Rubber Particles | 207.0 kg | 207.0 kg |
| Recycled Steel Fibres | 25.0 kg | 25.0 kg |
| Water | 300.5 kg | 320.0 kg |
| Super-plasticiser | 5.775 L | 5.775 L |
| Pre-treatment of rubber particles - Applied? | NO | YES |

as measured by the slump test, the common concrete consistence method [58]. The resulted slump test values were at 170 mm for the Treated and 175 mm for the Non-treated rubber mix, both classified under fresh concrete consistence category S4 [59]. Fig. 4 shows images of the slump test sample for both the Non-treated rubber mix (Fig. 4(a)) and the Treated rubber mix (Fig. 4 **(b)**).

### 2.4. Material characterization methods

#### 2.4.1. X-ray diffraction (XRD)

The WQD powder was examined using XRD, to determine its crystalline material composition. The XRD measurements were performed on a theta-theta, Rigaku Ultima IV diffractometer equipped with a Copper source, and a parabolic multilayer mirror for parallel X-ray beam, configured for Cu $K_a$ wavelength ($\lambda = 0{,}1541$ nm). All data were collected using 40 kV accelerating voltage and 40 mA current, while the diffracting intensity was measured by a scintillator. The incident and diffracted beam was conditioned with a divergent and a receiving slit, respectively, both of 0.5 mm opening.

#### 2.4.2. Scanning electron microscopy (SEM)/energy dispersive X-ray spectroscopy (EDX)

An EDX detector, integrated on a FEI Quanta 200 SEM was used to conduct elemental analysis of the WQD powder and fractured hardened SFRRC surfaces. Images of rubber particles before pre-treatment (as received) and after pre-treatment, as well as fractured surfaces of SFRRC samples from the Treated and Non-treated mixtures (2nd set) were obtained using the SEM. Before being inserted into the microscope, all samples were coated with a thin layer of silver (Ag) using an SC7640 High Resolution Sputter Coater, in order to increase sample conductivity and avoid surface charging effects. In addition to SEM imaging and EDX elemental analysis, in the case of hardened concrete surfaces, elemental EDX mapping was performed, indicating the elements present at specified areas on the SEM images.

### 2.5. Surface wettability testing

The wettability of a specific material thus its hydrophobicity or hydrophilicity is quantified by measuring the angle that forms between the material surface and a drop of liquid [60], referred to as the contact angle. Materials are considered hydrophobic when their contact angles are greater than 90 degrees, whereas materials are considered hydrophilic when contact angle measurements are lower than 90 degrees [60]. The surface wettability of rubber particles was determined before and after applying the WQD pre-treatment by the contact angle procedure using custom-made equipment while dropping water on the material surface using a syringe.

## 3. Results and discussion

The 7-day and 28-day compressive strength results are summarized in Table 3 and Table 4 for the 1st and 2nd set of mixtures respectively. Results from the 1st set of mixtures indicated that the WQD pre-treatment was able to increase the 28-day compressive strength of the Control mix by more than 300%, and surprisingly to a higher extend than replacing 3% of the original microsilica content with nanosilica. High variability in the Treated samples' compressive strengths led to the 2nd set of mixtures, where a direct comparison of using Non-treated versus Treated rubber particles was achieved. The results (Table 4) indicate an increase of 276% of the 28-day compressive strength and an increase of 282% of the 7-day compressive strength of the mixture.

The increased compressive strength is attributed to both reducing the porosity of the material as well as to enhanced ITZ achieved by the pre-treatment. The mechanism lies on converting the hydrophobic [31] rubber particles into hydrophilic, by applying the WQD pre-treatment. The wettability of rubber particles before and after applying the WQD pre-treatment was determined by the contact angle measurements. A water droplet on as-received rubber is shown in the macroscopic and magnified images of Fig. 5, indicating a contact angle greater than 90 degrees and indisputable hydrophobicity, as expected. The wettability of WQD pre-treated rubber was better captured by video; a time lapse of 4 images shown in Fig. 6 indicates the instantaneous absorption of the water droplet. The first image captured the moment the water was dropped on the surface of the pre-treated rubber (t = 0), indicating

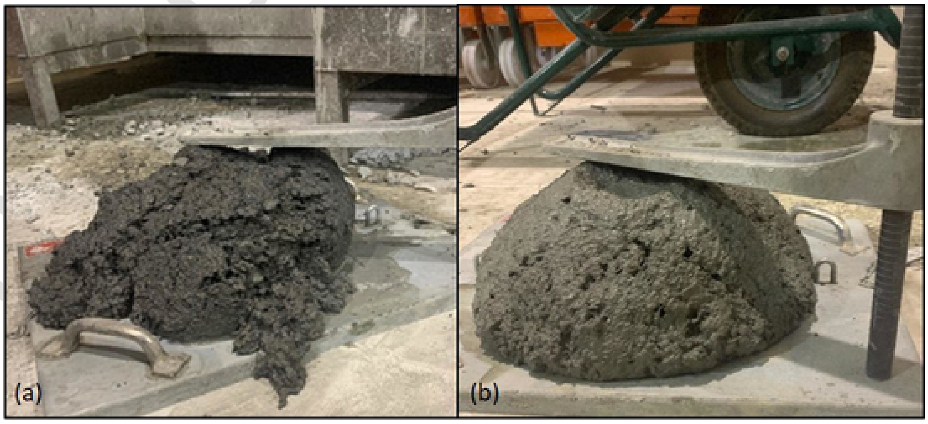

**Fig. 4.** Slump test images for (a) Non-treated mix and (b) Treated mix.

**Table 3**
Compressive Strength Results (1st set of mixtures).

| Mix ID | 7-day Compressive Strength (MPa) | | 28-day Compressive Strength (MPa) | |
|---|---|---|---|---|
| | Average | Std. Dev. | Average | Std. Dev. |
| Control | 1.63 | 0.06 | 2.17 | 0.06 |
| NanoP | 2.88 | 0.04 | 3.23 | 0.12 |
| NanoS | 4.47 | 0.01 | 4.74 | 0.12 |
| Treated | 2.48 | 0.09 | 6.97 | 1.99 |

**Table 4**
Compressive Strength Results (2nd set of mixtures).

| Mix ID | 7-day Compressive Strength (MPa) | | 28-day Compressive Strength (MPa) | |
|---|---|---|---|---|
| | Average | Std. Dev. | Average | Std. Dev. |
| Non-treated | 1.92 | 0.10 | 2.35 | 0.63 |
| Treated | 4.76 | 0.43 | 6.46 | 0.04 |

a fully hydrophilic surface with a contact angle clearly lower than 90 degrees.

The success of the WQD pre-treatment in eliminating the hydrophobicity of rubber particles is possibly a result of adherence of the fine WQD powder constituents to the rubber surface. The resulted XRD pattern of the investigated WQD powder sample is shown in Fig. 7, revealing the presence of the minerals tirodite, laumontite and andesine of the silicates group, as well as clinoclave, a chlorite group mineral. Zeolites L (based on Na) and Zeolites K-F (based on K) were also detected. Table 5 shows the EDX composition of the WQD powder, listing the elements identified and their corresponding atomic composition percentage within the sample. The elements detected through EDX have been instrumental in identifying the crystalline structures present in WQD probed through X-ray diffractograms. The clay minerals noted above and attached to the rubber particles through the pre-treatment process eliminate the inherent rubber hydrophobicity through their enhanced water adsorption capacity which subsequently reduces the overall void formation within SFRRC.

In addition to analysing the WQD powder sample, rubber particles were also examined, both as received and WQD pre-treated. Macroscopic and magnified SEM images of as received versus WQD pre-treated rubber particles are shown in Fig. 8, indicating the effects of pre-treatment on changing the surface appearance. Further analysis of the rubber particle surface chemistry was obtained by EDX composition and elemental analysis of specific points on the magnified SEM images. The EDX composition and elemental analysis of rubber as received and pre-treated with WQD are shown in Table 5, demonstrating the capacity of the pre-treatment method to generate a stable WQD coating on the rubber particles.

By eliminating the hydrophobicity of rubber, the WQD pre-treatment was able to significantly reduce the SFRRC porosity, obvious with the naked eye on the hardened SFRRC cube specimen surfaces shown in Fig. 9. The air void reduction is also visible when observing the material internally, as indicated in macroscopic and magnified SEM images of fractured SFRRC surfaces (Fig. 10), where (a), (c) Non-treated and (b), (d) Treated SFRRC mixtures of the 2nd set are compared.

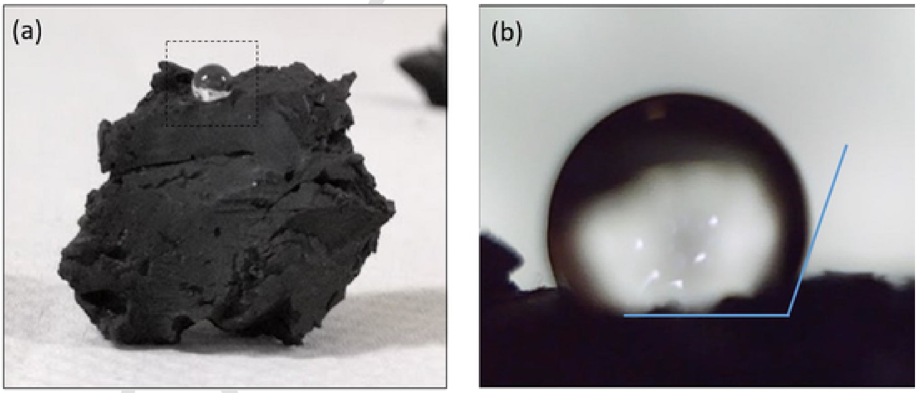

**Fig. 5.** (a) Water droplet on a non-treated rubber particle. (b) Magnified version of the water droplet demonstrating the contact angle measurement.

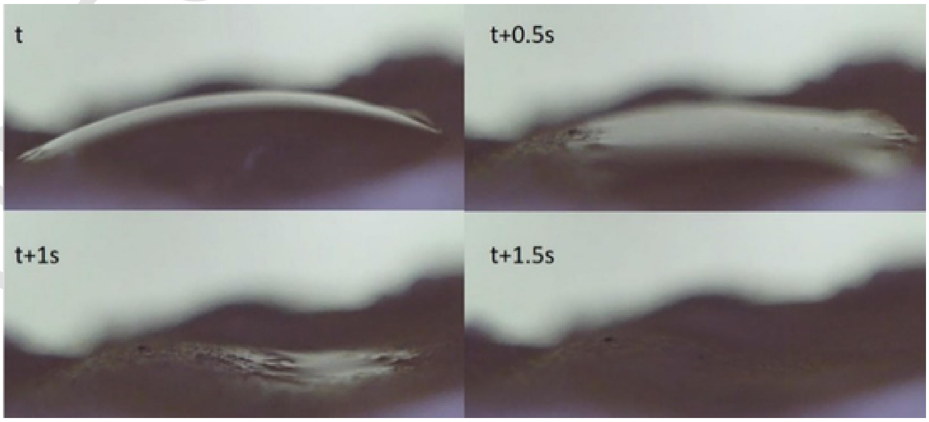

**Fig. 6.** Water droplet on WQD pre-treated rubber particle.

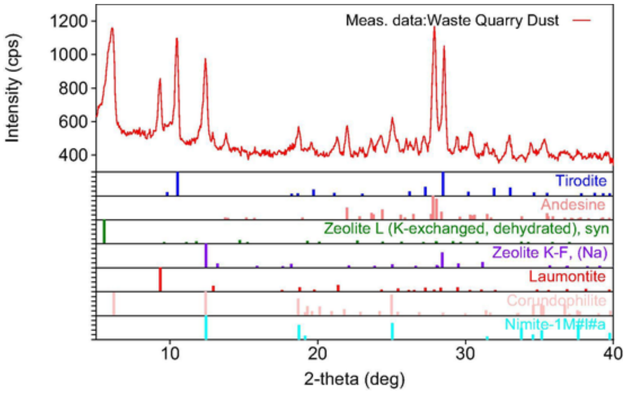

**Fig. 7.** WQD powder experimental XRD pattern.

**Table 5**
EDX Composition of waste quarry dust and rubber particles, as received and pre-treated with waste quarry dust.

| | Waste Quarry Dust (WQD) | Rubber Particles (RP) | WQD Pre-treated RP |
|---|---|---|---|
| Element | Composition at % | | |
| C | 19.6 | 91.2 | 12.9 |
| O | 46.4 | 5.9 | 30.0 |
| Na | 2.4 | 0.8 | 1.5 |
| Mg | 3.4 | – | 3.5 |
| Al | 6.0 | 0.2 | 9.0 |
| Si | 16.0 | 0.2 | 22.6 |
| S | 0.1 | 1.2 | – |
| Ca | 2.8 | – | 10.8 |
| Ti | 0.2 | – | 0.3 |
| Fe | 3.0 | – | 9.4 |
| Zn | – | 0.4 | – |

Reduction of the overall air void formation due to successful elimination of rubber particle hydrophobicity is also evident when comparing SFRRC sample densities of Treated to Non-treated mix. Table 6 summarises the hardened SFRRC sample densities, indicating denser Treated mix samples compared to Non-treated mix samples, by 9.7% and 7.8% for 7-day and 28-day hardened SFRRC samples respectively.

Analysis of the rubber-cement paste interface was conducted to observe the effects of WQD pre-treatment on enhancing the ITZ of rubberized concrete. Fractured SFRRC surfaces from both Treated and Non-treated mixture samples were analysed by SEM/EDX, with images taken in a wide range of magnification scales to illustrate the macroscopic appearance of the interface areas and detect micro and nanoscale constituents of the material. Elemental maps were used to identify rubber and cement paste areas as shown in Fig. 11. The rubber-cement paste interface shown in Fig. 11 (b), (d) (Treated), appears enhanced compared to that of Fig. 11 (a), (c) (Non-treated), with visible reduction in the air voids and improved bonding observed.

A closer look in Fig. 11 (a), that represents the rubber-cement paste interface of a fractured concrete surface from a Treated mix sample where the rubber particles used were pre-treated, clearly indicates the reduction of air voids at the rubber-cement paste interface, achieved by the WQD pre-treatment. In contrast, a closer look into a SEM image of the fractured hardened SFRRC mixture that included non-treated rubber particles (Fig. 11 (d)) shows a more porous arrangement with an abundance of needle-like crystals, identified as ettringite, in agreement with previous research [33] that notes the abundance of ettringite in concrete with high rubber aggregate content. It should be noted that no abundance of needle-like crystals was observed in the case of the Treated mix.

## 4. Conclusions

The pre-treatment is successful at reversing hydrophobic rubber surface into hydrophilic, as indicated by contact angle measurement images . This modification further enhances rubber particle behaviour, when used as aggregate replacement in concrete and mixed within a cementitious paste. By eliminating hydrophobicity, fluid repelling by the rubber particles and consequent formation of excessive air pores do not occur during mixing; thus, significantly reducing rubberized concrete porosity and increasing the material density. The success of the WQD pre-treatment in reducing the hydrophobicity of rubber particles is possibly due to adherence of the WQD constituents onto the rubber surface. Further study of this mechanism is required to obtain clear understanding. Observations on fractured surfaces of hardened SFRRC samples and density measurements indicate an obvious reduction in the porosity of the mixture that included pre-treated rubber particles. SEM images further demonstrate this effect and provide interesting close-ups of rubber-cement paste interface locations. Comparing the magnified images of SFRRC with non-treated versus SFRRC with WQD pre-treated rubber particles, it is observed that not only the porosity of the material is reduced, but a significant reduction of the gaps observed at the ITZ is also achieved. For the case of SFRRC where non-treated rubber particles were used, an abundance of Ettringite is indicated, in agreement with previous research [33], with the interesting addition that this behaviour is not observed in the SEM images of SFRRC made with pre-treated rubber. Further study focusing on hydration products is necessary to provide further statements. Most importantly, the recommended pre-treatment was able to increase both the 7- and 28-day compressive strengths of the SFRRC mixture investigated by 282% and 276%, respectively. While rubberized concrete suffers significant compressive strength reduction with inclusion of rubber in replacement to mineral aggregate, a novel technique, using another waste material has been proven effective in reducing this detrimental effect. Following this proof of concept study, repeatability testing is recommended to verify the effectiveness of the method. Furthermore, studying the effects of this novel pre-treatment on the modulus of elasticity of the material, tensile strength as well as damping and energy absorption capacities of rubberized concrete is recommended, specifically for mixtures with such high rubber content, aimed for the development of forgiving infrastructure applications such as road safety barriers.

## CRediT authorship contribution statement

**Thomaida Polydorou:** Conceptualization, Methodology, Investigation, Writing - original draft. **Georgios Constantinides:** Conceptualization, Methodology, Investigation, Writing - review & editing. **Kyriacos Neocleous:** Conceptualization, Methodology, Investigation, Writing - review & editing. **Nicholas Kyriakides:** Methodology, Investigation, Writing - review & editing. **Loukas Koutsokeras:** Investigation. **Christis Chrysostomou:** Supervision. **Diofantos Hadjimitsis:** Project administration, Funding acquisition.

## Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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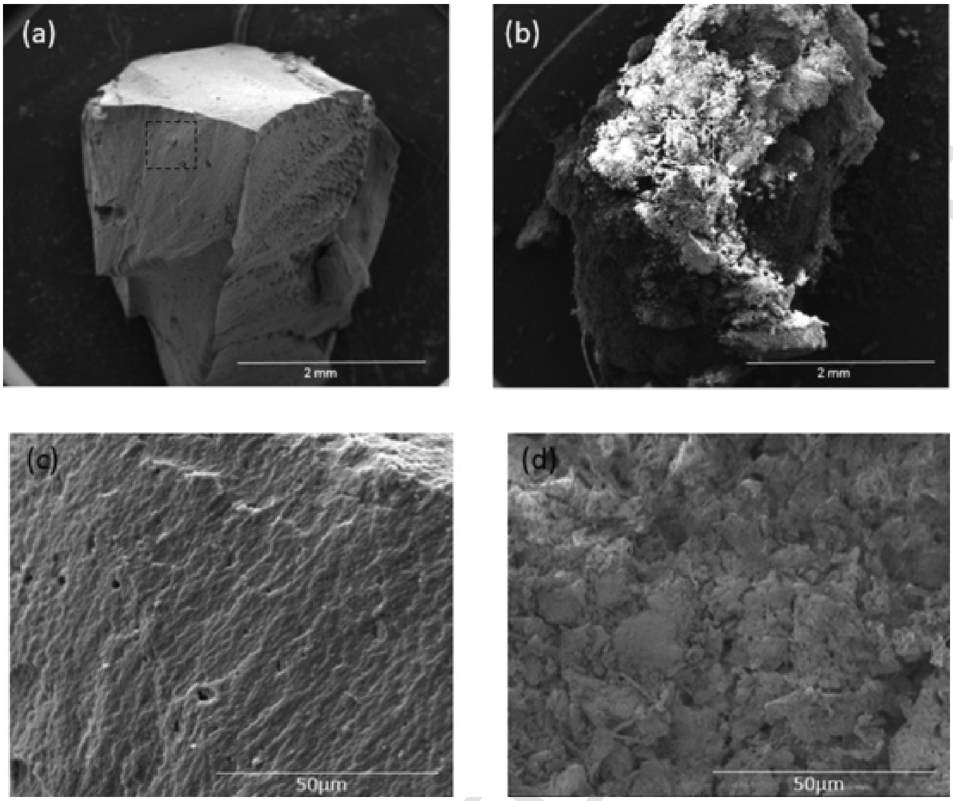

**Fig. 8.** Macroscopic and magnified images of rubber particles (a), (c) as received from tyre recycling plant and (b), (d) pre-treated with WQD.

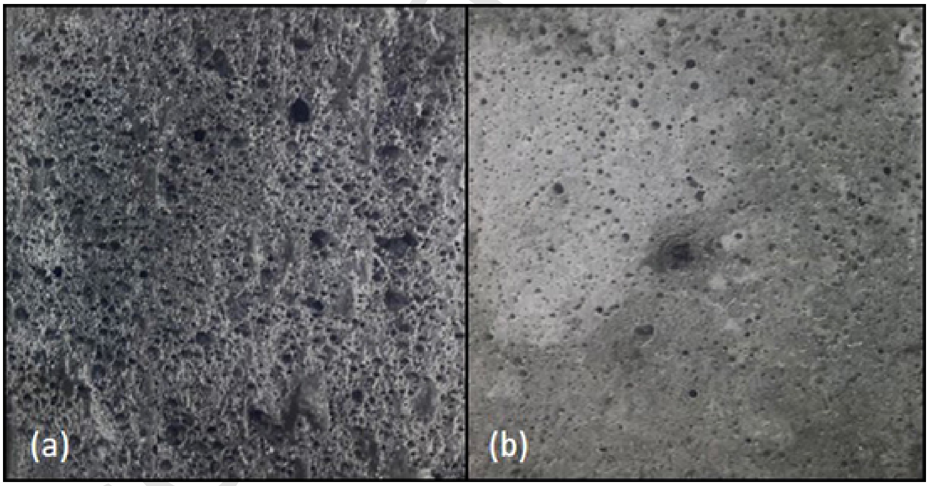

**Fig. 9.** Macroscopic images of 7-day cured SFRRC cube specimen surfaces (a) Non-treated mix and (b) Treated mix.

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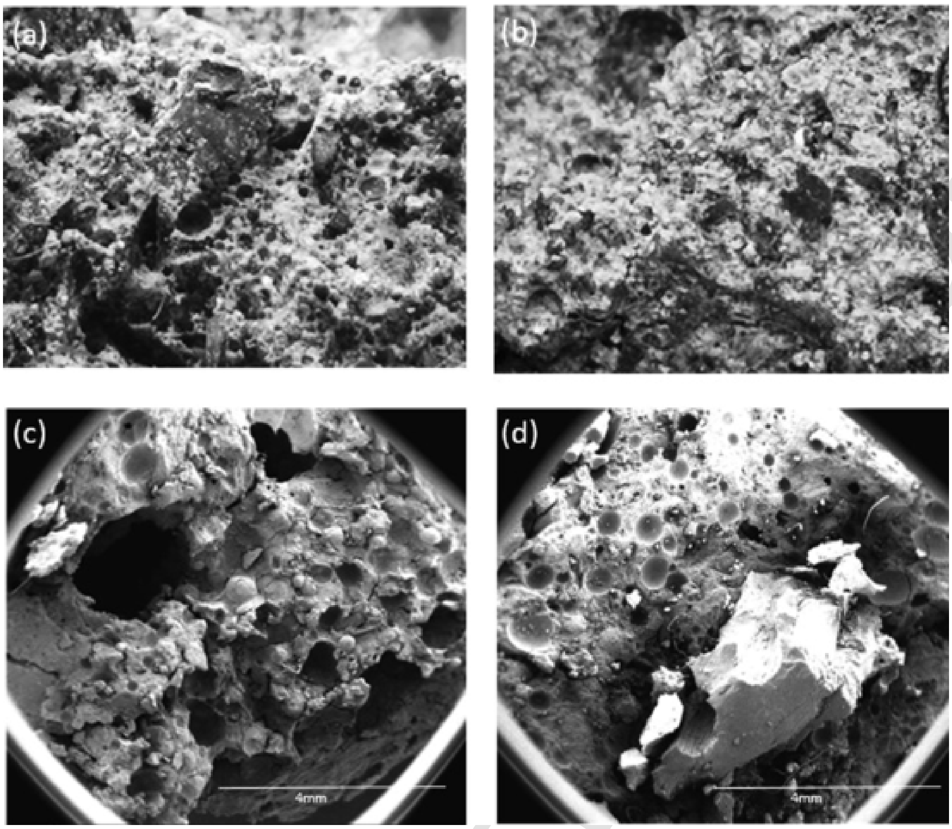

**Fig. 10.** Macroscopic and magnified images of fractured SFRRC surfaces (a), (c) Non-treated mix and (b), (d) Treated mix.

**Table 6**
Hardened SFRRC sample densities.

| Mix ID | 7-day hardened SFRRC sample density (kg/m $^3$) | | 28-day hardened SFRRC sample density (kg/m $^3$) | |
|---|---|---|---|---|
| | Average | Std. Dev. | Average | Std. Dev. |
| Non-treated | 1458.47 | 30.55 | 1488.20 | 21.48 |
| Treated | 1600.01 | 10.83 | 1604.44 | 10.06 |

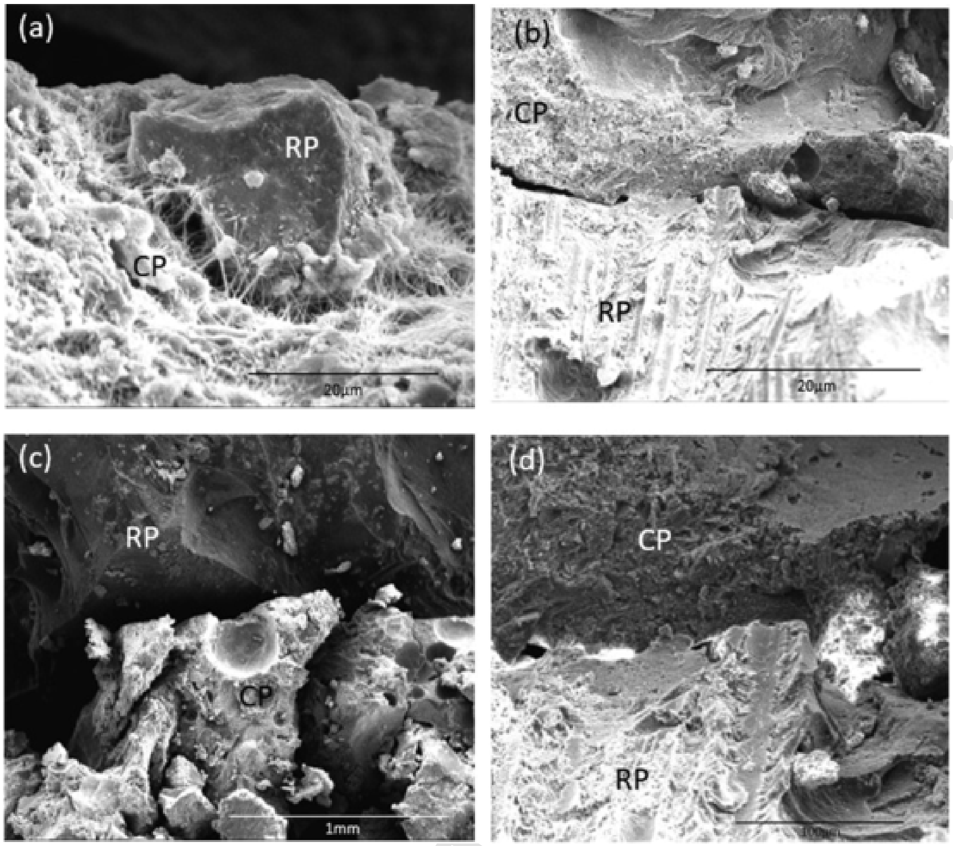

**Fig. 11.** SEM images of the rubber-cement paste interface from fractured surfaces of the non-treated mix (a), (c) and treated mix (b), (d). Rubber particles (RP) and cement paste (CP) areas are identified through EDX elemental mapping and noted in the images.

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