RUBBERISED CONCRETE REFINEMENT BY CEMENT SUBSTITUTION AND RUBBER PARTICLE PRETREATMENT

Dr. Thomaida Polydorou (1), Dr. Kyriacos Neocleous (1), Dr. Loukas Koutsokeras (2), Dr. Georgios Constantinides (2), Dr. Nicholas Kyriakides (1), Prof. Kypros Pilakoutas (3) and Prof. Diofantos Hadjimitsis (1)

(1) Department of Civil Engineering and Geomatics, Cyprus University of Technology, Cyprus

(2) Department of Mechanical Engineering and Materials Science and Engineering, Cyprus University of Technology, Cyprus

(3) Department of Civil and Structural Engineering, The University of Sheffield & Department of Civil Engineering and Geomatics, Cyprus University of Technology (Visiting Professor)

Abstract

Re-use of End-of-life tyre components into concrete is a viable solution to the environmental issue of tyre waste that can result in promising, high added value concrete applications. End-of-life tyre particles are added to conventional concrete by replacement of a percentage of its aggregate content, improving concrete deformability. The material is currently in the initial stages of research for structural applications, with weakness observed over the interfacial transition zone between the rubber and cement paste.

This paper examines the potential of cement substitution by a combination of micro and nanoscale silica particles as well as the effect of rubber pre-treatment by coating with a Diabase quarry dust slurry for surface modification; aiming to refine the rubber/cement paste interface in rubberised concrete, thus improving its compressive strength and consequently structural performance.

The effectiveness of cement substitution and rubber treatment methods is measured by comparison of the samples compressive strength at 7 and 28 days and detected in microscopic observations of hardened rubberised concrete samples. It is indicated the rubber pre coating method investigated is a promising procedure, proven to be effective in reducing the porosity at the rubber-cement paste interface and increasing the 28-day concrete compressive strength more than twice.

Keywords: rubberised concrete, fibre reinforcement, end-of-life tyres, circular economy, cement substitution.

1. INTRODUCTION

While the world is turning to a more circular economy [1], researchers are investigating the re-use of End-of-life tyre components in high added value applications. A circular economy promotes reusing products, rather than scrapping what was formerly considered waste and then obtaining new resources. In such an economy, clothes, scrap metal and obsolete electronics, are not considered waste, but are instead returned to the economy or used more efficiently [1], [2].

Management of End-of-Life tyres is a major environmental concern in many countries; stockpiling of End-of-Life tyres is not only aesthetically unpleasing but also dangerous and in Europe has been outlawed through the implementation of EU Waste Legislation [2], [3]. Recycling of tyre rubber in construction materials and road furniture is an ideal solution to a significant environmental, health, and aesthetic problem [4], [5], [6].



Figure 1-1: Recycled Tyre Rubber Particles

Reuse of End-of-life tyre components (e.g. rubber) into concrete, the second most consumed material in the world after water [7], is considered one of the most viable solutions for this dangerous waste issue; not only eliminating large amounts of the waste rubber, but also providing a unique property to an otherwise non-deformable material. Conventional concrete lacks elasticity or flexibility unless reinforced with steel [8].

While Rubberised Concrete is a promising material, challenges regarding its performance have not been addressed sufficiently [9], [10]. High rubber content can decrease concrete compressive strength significantly thus thoughtful mix design based on the application is crucial for an effective material. The decrease in compressive strength with increasing rubber content is attributed to the higher Poisson's ratio of the rubber, compared to the replaced mineral aggregates, as well as to the poor bonding observed between the rubber particles and the cement paste, also referred to as weak Interfacial Transition Zone (ITZ). The bonding between rubber particles and cement paste at their ITZ was studied through Scanning Electron Microscopy (SEM) images, where gaps due to lack of bonding were visible and limited hydration products were observed around the rubber particles [11]. The higher porosity observed at the rubber-cement paste interface is also attributed to the hydrophobic nature of the rubber particles. In the similar case of recycled aggregates (RA), a method that was effective in improving the interfacial transition zone leading to higher compressive strengths was aggregate coating with very fine materials [12].

There are no methods established yet for the characterisation of recycled rubber properties; thus making it difficult to optimise the behaviour of rubberised concrete. Providing an optimum

gradation of rubber particles is shown to improve rubberised concrete compressive strength due to better packing of the mixture contents. In addition, admixtures such as plasticizers and superplasticisers are used to achieve better consolidation of the mixture at predetermined water to binder ratios to control compressive strength. Even though previous research agrees that increasing rubber content results to a significant decrease in compressive strength, some argue that the rubber bonds well to the cement matrix [13], [14], [15].

Research on rubberised concrete is currently contradictory since rubber particles obtained from different recycling plants or even the same plant, vary when it comes to contamination levels (e.g. rubber dust, textile/polymeric fibre) and surface roughness. Rubber extraction and size-reduction methods are not consistent between tyre recycling plants. In addition to the traditional ambient-temperature mechanical treatment methods, water-jet cutting methods and de-vulcanization of rubber particles are also used in some tyre recycling plants worldwide, stating that de-vulcanized rubber surface is highly advantageous compared to vulcanised rubber, and can enhance the mechanical properties of rubberised concrete [16].

Mixture workability is also a primary issue in rubberised concrete, with difficulties observed due to the relatively low density of the rubber particles compared to the included natural aggregates and cement [11]. By sufficient consolidation and release of the mixture's entrapped air, rubberised concrete is able to attain satisfactory homogeneity.

In addition to rubber, tyre recycling also yields recycled tyre steel wires (Figure 1-2), which have limited alternative applications for their use; the most common one being scrap feed in steel making. It is rather preferred that recycled tyre steel wires are reused in high-value applications that can benefit from the materials' exceptional physical properties [17], [6].



Figure 1-2 : Recycled Tyre Steel Fibres

Extensive work has been undertaken on the use of recycled tyre steel fibres in concrete, where studies have compared sustainable hybrid fibre–reinforced concrete (SHFRC) to ordinary fibre-reinforced concrete with only manufactured fibres and demonstrated that SHFRC can have equal or better properties than ordinary fibre reinforced concrete [18], [19].

Methods for extracting, cleaning and sorting the recycled tyre steel tyre fibre (RTSF) were developed and optimised [20], resulting to the assembly of a RTSF product of known properties [3], [21], [19]. Unlike rebar and other types of reinforcement, RTSF allows for efficient microcrack control well before large cracks form. The unique characteristics of RTSF and the massive fibre count increase the local toughness of concrete and set new standards for sustainability [22], [23].

This research paper examines the possibilities of improving the mechanical properties of steel fibre reinforced rubberised concrete by rubber treatment with waste quarry dust paste and cement replacement by both micro and nano scale silica particles.

2. MATERIALS AND METHODS

A steel fibre reinforced concrete mix with 60% aggregate replacement by rubber particles was optimised for the development of a Steel Fibre Reinforced Rubberised Concrete Barrier, during the experimental testing rounds of the research project SAFER, funded under the Horizon 2020 Marie Skłodowska-Curie Actions, Grant Agreement No 748600. The SAFER Mix development has taken into consideration the findings of the collaborative project 'Anagennisi', funded under FP7 Grant Agreement No 603722.

To build on the investigation by the 'Anagennisi' team on enhancing the compressive strength of rubberised concrete, this research examines possible methods that could aid in improving the material compressive strength of the SAFER Steel Fibre Reinforced Rubberised Concrete Mix (S-SFRRC), by experimental examination of 3 mixture modifications.

The mixture modifications investigated by this study are summarised in Table 2.1. Mix A is a modified version of the optimised SAFER project mixture, adapted for the needs of this specific study. Mix B varies compared to Mix A only by the fact that a simple pre-treatment technique was applied to all rubber particles of the mixture, 1 day before casting and after the total amount of rubber for the Mix was weighed out and rubber particles were all blended together. The pre-treatment applied to the rubber particles of Mix B involved immersing them into a slurry made of waste diabase quarry dust from a local source and water for 1 hour. After mixing to ensure that all rubber particles' surfaces were covered by the slurry, the particles were scooped out and let dry on a flat surface for 24 hours, resulting to a thin coating on the rubber particles' surfaces.

Mix C and Mix D of this study examine the effect of replacing 3% of the microsilica content of Mix A with 15-20mm diameter SiO_2 particles, or nanosilica. The particles used in Mix C were nearly nonporous amorphous and spherical (S-type) but the particles used in Mix D were porous and amorphous (P-type).

As mentioned in the preceding paragraphs, the original SAFER project optimised mixture was not suitable for this study, due to the fact that when 3% of its microsilica content was replaced by nanosilica, the mixture became unworkable therefore additional water and superplasticiser were used to accommodate this issue. Accordingly, all 4 mixtures in this study included the additional amount of water and superplasticiser required to make Mix C and D workable, to ensure a fair comparison between the compressive strengths of the 4 mixture samples. It should be noted that the original SAFER mixture water to binder (w/b) ratio is 0.45 but the mixtures in this study all have a w/b ratio of 0.56. Table 2.1 shows the constituents and quantities used per cubic meter of material, for each of the 4 investigated mixtures.

The SAFER Steel Fibre Reinforced Rubberised Concrete Mix has 60% (by volume) of its natural aggregates replaced by rubber particles of equal size, keeping a fine aggregate to coarse aggregate ratio of 1.22.

The mix included rubber particles from 3 different sources coming from 3 different countries in Europe (i.e. Croatia, Cyprus and the UK) with no material properties specified. A representative sample of all types of rubber particles from the sources used in this study was tested for its apparent particle density, following EN 1097-6 (CEN 2013). The average Specific Gravity of the representative rubber particle sample used in this mix was determined to be 0.8. For this research, liquid polycarboxilic polymer based superplasticiser conforming to EN934-2:2009 was EN 197-1 portland cement CEM I 52,5N was used.

The Recycled Tyre Steel Fibres (RTSF) used in this study are commercially cleaned, sorted and evaluated. The product properties are as follows; Tensile strength: $2,560 \pm 550$ MPa Young's Modulus: $200,000 \pm XXX$ MPa (Twincon Ltd, 2018).

Microsilica conforming to ASTM C1240 requirements and also fulfilling the requirement of Table ZA.1 in EN13263-1:2005+A1:2009 for Silica Fume-Class 1 was used in this study. In modified mixtures C and D, 3% of the Microsilica content was replaced by Nanosilica. The S-type nanosilica used in Mix C is Silicon Oxide Nanopowder / SiO2 Nanoparticles (SiOx, 99.5⁺%, 15-20nm, S-type, Spherical, Nonporous and amorphous). The P-type nanosilica used in Mix D is Silicon Oxide Nanopowder / SiO2 Nanoparticles (SiOx, 99.5⁺%, 15-20nm, Porous and amorphous).

A 60-Litre capacity drum mixer was used to make the specimens. First, all aggregates were added into the mixer and blended for 2-3 minutes before adding half of the mixing water; mixing was carried out for 2 minutes. The binders were added subsequently, followed by the rest of the water and admixture liquid. The steel fibres were added last, during the final 3 minutes of mixing. The Steel Fibre Reinforced Rubberised Concrete samples were cast and consolidated using a vibrating table. After 48 hours in their moulds, the samples were de-moulded and placed in a water tank to cure until tested. Compressive strength was tested at 7 and 28 days for all specimens and at 90 days for modified mixtures C and D only. Compressive strength testing was performed after allowing the wet specimens to dry for 24 hours.

Mixture Constituent	Mix ID-A	Mix ID-B	Mix ID-C	Mix ID-D
Cement CEM I 52,5N	400.0 kg/m^3	400.0 kg/m^3	400.0 kg/m^3	400.0 kg/m^3
Silica Fume (Micro-silica)	100.0 kg/m^3	100.0 kg/m^3	97.0 kg/m^3	97.0 kg/m^3
P-type Nanosilica particles (15-	-	-	-	3.0 kg/m^3
20nm)				
S-type Nanosilica particles (15-	-	-	3.0 kg/m^3	-
20nm)				
Fine Crushed Aggregate (0-4mm)	310.5 kg/m ³	310.5 kg/m ³	310.5 kg/m ³	310.5 kg/m ³
Coarse Crushed Aggregate (4-	378.0 kg/m ³	378.0 kg/m^3	378.0 kg/m^3	378.0 kg/m ³
20mm)				
Fine Rubber Particles	169.7 kg/m ³	169.7 kg/m ³	169.7 kg/m ³	169.7 kg/m ³
Coarse Rubber Particles	207.0 kg/m^3	207.0 kg/m^3	207.0 kg/m^3	207.0 kg/m^3
Rubber Particle Treatment	-	\checkmark	-	-
Recycled Steel Fibres	25.0 kg/m^3	25.0 kg/m^3	25.0 kg/m^3	25.0 kg/m^3
Water	280.0 kg/m^3	280.0 kg/m^3	280.0 kg/m^3	280.0 kg/m^3
Super-plasticiser	$4.875 (L/m^3)$	4.875 (L/m ³)	4.875 (L/m ³)	4.875 (L/m ³)

Table 2.1 : Mixture Constituents

3. **RESULTS AND DISCUSSION**

The average compressive strength and standard deviation values for each of the 4 Mixtures investigated by this study at 7, 28 and 90 days as applies, are listed in Table 3.1.

	7-day Strength (MPa)		28-day Strength (MPa)		90-day Strength (MPa)	
	Average	Standard	Average	Standard	Average	Standard
	_	Deviation	_	Deviation	_	Deviation
Mix A	1.63	0.06	2.17	0.06	-	-
Mix B	2.48	0.09	6.97	1.99	-	-
Mix C	4.47	0.01	4.74	0.12	4.80	0.08
Mix D	2.88	0.04	3.23	0.12	3.25	0.01

Table 3.1 : Compressive Strength Average and Standard Deviation Values

The compressive strength results indicate that both cement substitution by both silica fume (microsilica) and S or P type nanosilica, as well as the rubber pre-treatment method investigated in this research were all effective in increasing the mixture compressive strength. As expected, a significant increase of the early compressive strength is achieved by including a small amount of spherical SiO₂ nano particles in the mixture, specifically a 174% increase of the average 7-day compressive strength with the substitution of 3% of the SiO₂ micro size particles (silica fume or microsilica) by non-porous spherical SiO₂ nano size particles. Where substituted by porous SiO₂ nano size particles (Mix D), the 7-day compressive strength increase was limited to 77%. A 52% increase of the 7-day compressive strength is observed with the pre-treatment method investigated (Mix B), where unlike the cases of Mix C and D, there was no cement replacement by an expensive nanomaterial but instead, a simple pre-treatment using waste dust was applied on the rubber particles of the mixture.

At 28 days, the steel fibre reinforced rubberised concrete compressive strengths of all trial mixtures increased further, with no significant strength gains in the cases of cement replacement by combined micro and nano SiO₂ (Mix C, Mix D). Inversely, the average compressive strength of the samples where the rubber particles went through the pre-treatment increased by 181% during the additional 21 days of curing (Mix B) and having a 221% higher 28-day compressive strength compared to Mix A, between which no variant exists other than the rubber pre-treatment. It is speculated that the rubber coating achieved by pre-treatment with the waste diabase quarry dust was able to reduce the hydrophobicity of the rubber particles, thus eliminating the resulting porosity in the mixture. In contrast with the values obtained for the rest of the mixtures, a high variability was observed in the case where pre treated rubber particles were used. The variability in compressive strength in this case is attributed to the fact that the slurry coating saturation state was inconsistent within the sample range.

When comparing Mix C and Mix D, where the only difference between the two is the type of SiO_2 particles used; non-porous in Mix C and porous in Mix D, the higher compressive strength attained by Mix C samples can be explained by the contribution of density from the non-porous SiO_2 nanoparticles to the total mix density.

The apparent densities of the hardened concrete samples, listed in Table 3.2, relate to the respective compressive strengths reported in Table 3.1, suggesting that the main factor of strength improvement is the densification of the cement paste matrix.

Apparent Density (kg/m ³)	Mix A	Mix B	Mix C	Mix D
7-day cured samples	1164	1335	1386	1272
28-day cured samples	1328	1618	1411	1290

Table 3.2 : Apparent Density of Hardened Concrete Samples

This is further enforced through Scanning Electron Microscopy (SEM) images of fractured surfaces. Figure 3.1 shows images of the non-treated (Mix A) vs. the treated (Mix B) rubber surfaces suggesting that the treatment process results in a significant reduction in the volume of pores present in the microstructure. While this is in agreement with the observed density increase, it is not yet clear whether the porosity reduction relates to an enhanced hydrophilicity of the rubber particles induced by the treatment process. Furthermore, other mechanisms including the formation of cement hydration products as well as chemical interactions of the mix constituents with the waste dust used for the slurry coating need to be investigated.



Figure 3-1 : Scanning Electron Microscopy images of fractured surfaces of hardened SFRRC; Mix A sample (A) and Mix B sample (B)

4. CONCLUSIONS

The rubber particle treatment method investigated by this study appears to be very promising. A significant increase in the compressive strength was achieved by a simple rubber pre-treatment method using diabase quarry dust which is considered waste. Substitution of the mixture's microsilica content with non-porous nanosilica increased the material compressive strength but not to the extend where the cost of the nanomaterial is justified. Further investigation should be undertaken, to determine the chemical characteristics of the quarry dust and the effects it has on the rubber particles surface properties as well as the chemical interaction of the dust particles during the cement hydration process. In addition, the treatment process should be optimized and standardized to reduce the variability observed in the compressive strengths of samples which included the slurry treated rubber particles.

ACKNOWLEDGEMENTS

This research has received funding from the European Union's Horizon 2020 research and innovation programme under the Marie Skłodowska-Curie grant agreement No 748600.

REFERENCES

- [1] United Nations (2018), United Nations Conference on Trade and Development No.61.
- [2] Directive 2008/98/EC (2008), Official Journal of the European Union, Directive 2008/98/EC of the European Parliament and Council on Waste and Repealing Certain Directives, Brussels, Belgium.
- [3] Pilakoutas, K., Neocleous, K. & Tlemat, H. (2004), "Reuse of tyre steel fibres as concrete reinforcement", Engineering Sustainability 157 (ES3), 131-138.
- [4] Khaloo A. R., Dehestani, M. & Rahmatabadi, P. (2008), "Mechanical properties of concrete containing a high volume of tire–rubber particles", Waste Management 28 (2008) 2472–2482.
- [5] Khatib Z. K., & Bayomy, F. M. (1999), "Rubberized Portland cement concrete", Journal of Materials in Civil Engineering, 1999, 11(3): 206-213.
- [6] European Tyre Recycling Association (ETRA), <u>http://www.etra-eu.org/joomla/projects</u>, 2016.
- [7] Gagg, Colin (2014), "Cement and concrete as an engineering material: An historic appraisal and case study analysis", Engineering Failure Analysis 40, 114-140.
- [8] Sealy, Cordelia (2018), "Nanoscale order brings flexibility to concrete", Nano Today 18(2018)1–7.
- [9] Elchalakani, M. (2015), "High strength rubberized concrete containing silica fume for the construction of sustainable road side barriers", Structures 1 (2015) 20–38.
- [10] Khalil, E., Abd-Elmohsen, M. & Anwar, A. M., (2015), "Impact Resistance of Rubberized Self-Compacting Concrete", Water Science 29 (2015) 45-53.
- [11] Raffoul, S., Garcia, R., Pilakoutas, K., Guadagnini, M. & Medina, N. F. (2016), "Optimisation of rubberised concrete with high rubber content: An experimental investigation", Construction and Building Materials 124 (2016) 391-404.
- [12] Younis, K. H. & Pilakoutas, K. (2013), "Strength prediction model and methods for improving recycled aggregate concrete", Construction and Building Materials 49 (2013) 688-701.
- [13] Bignozzi, M. C. & Sandrolini, F. (2006), "Tyre rubber waste recycling in self-compacting concrete", Cem. Concr. Res. 36 (4) (2006) 735–739.
- [14] Benazzouk A., Douzane, O., Langlet T., Mezreb K., Roucoult J. M. & Quéneudec M. (2007), "Physico-mechanical properties and water absorption of cement composite containing shredded rubber wastes", Cem. Concr. Compos. 29 (10) (2007) 732–740.
- [15] Alsaif, A., Koutas, L., Bernal, S. A., Guadagnini, M. & Pilakoutas, K. (2018), "Mechanical performance of steel fibre reinforced rubberised concrete for flexible concrete pavements", Construction and Building Materials 172 (2018) 533-543.
- [16] Rubber Products Ltd (2016), "NRG Presentation 2016", https://www.rubber-products.net
- [17] Tlemat, H., Pilakoutas, K. & Neocleous, K. (2006), "Stress-strain characteristics of SFRC using recycled fibres", Materials and Structures 39 (3):365-377, 2006.
- [18] Baricevic, Ana & Bjegović, Dubravka & Skazlic, Marijan. (2017). Hybrid Fiber–Reinforced Concrete with Unsorted Recycled-Tire Steel Fibers. Journal of Materials in Civil Engineering. 29. 06017005. 10.1061/(ASCE)MT.1943-5533.0001906.
- [19] Hu H, Papastergiou P, Angelakopoulos H, Guadagnini M & Pilakoutas K (2018), "Mechanical properties of SFRC using blended Recycled Tyre Steel Cords (RTSC) and Recycled Tyre Steel Fibres (RTSF)", Construction and Building Materials, 187, 553-564.
- [20] Angelakopoulos H. & Waldron P., "TWINCLETOES-Tyre Wire in Concrete Leading to Environmental Sustainability", Layman's report, Twincon Ltd, Sheffield, UK (2015).
- [21] Graeff AG, Pilakoutas K, Neocleous K & Peres MVNN (2012), "Fatigue resistance and cracking mechanism of concrete pavements reinforced with recycled steel fibres recovered from postconsumer tyres", Engineering Structures, 45, 385-395.
- [22] Twincon Ltd., (2018), "Technical Data Sheet Product: RTSF", www.twincon.co.uk.
- [23] Pilakoutas, K. (2016), "Mechanical behaviour of steel fibre-reinforced rubberized concrete: preliminary results of the FP7 Anagennisi project, Personal Communication, June 2016, Limassol, Cyprus".