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NANOMATERIALS IN RUBBERISED CONCRETE: A REVIEW OF LATEST RESEARCH TRENDS

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ABSTRACT

The global push for sustainability is influencing all sectors, including construction, where the circular economy concept is gaining prominence by recycling waste materials. Waste tyres have captured significant attention from researchers as a resource for producing crumb rubber (CR), which can substitute for traditional fine aggregates in concrete. This substitution offers increased toughness, energy absorption, flexibility, and sound and heat insulation. However, the trade-off is a reduction in mechanical strength owing to inadequate bonding between CR and cement paste, attributed to the hydrophobic nature and softness of rubber particles compared to sand. Additionally, using CR increases matrix porosity. Various efforts have been made to address these challenges, and incorporating nanomaterials is one promising approach. This paper provides a concise review of the application of nanomaterials in rubberised concrete (RC) to mitigate the issues associated with CR in cementitious composites. It aims to serve as a quick guide for researchers interested in exploring nanomaterials in RC, offering insights into recent trends and the types of nanomaterials utilised in current research. The most commonly used nanomaterials in RC include graphene oxide (GO), graphene nanoplatelets (GNPs), nano silica (NS), carbon nanotubes (CNTs), nano iron oxide (NIO), and nano titanium dioxide. Due to differences in size and morphology, these nanomaterials enhance RC through distinct mechanisms. Zero-dimensional (0D) nanomaterials like NS act as nanofillers and reactive pozzolans, while one-dimensional (1D) nanomaterials such as CNTs can fill nanovoids and bridge microcracks. GO, a highly reactive two-dimensional (2D) nanomaterial, not only promotes cement hydration by acting as nucleation sites but also bridges across cracks, preventing their propagation. Additional nanomaterials that warrant further exploration in RC include carbon nanofibers, carbon black, nano calcium carbonate ($CaCO_3$), and innovative waste-based nanomaterials.

Keywords: *Carbon nanotubes, crumb rubber, fine aggregate, graphene oxide, nanotechnology, recycling*

INTRODUCTION

In recent years, the construction industry has increasingly focused on sustainable and environmentally friendly practices. One such approach is the incorporation of waste materials into construction products. Crumb rubber, derived from recycled tyres, has garnered attention for its potential use in concrete, known as rubberised concrete (RC). This innovative material offers the dual benefits of recycling waste tyres and enhancing specific properties of concrete [1]-[3].

CR is created by shredding and grinding scrap tyres into small particles. These particles are then incorporated into concrete as a partial substitute for fine aggregates [4]. Rubberised concrete has been proposed for various applications, including road construction [5]-[8], noise barriers [9]-[10], and non-structural elements [11], due to its potential for improved toughness, ductility, and energy absorption [12]-[14]. Nevertheless, the integration of crumb rubber into concrete poses notable challenges. The insufficient bonding between the hydrophobic rubber particles and the hydrophilic cement matrix leads to poor interfacial adhesion. This weak bonding reduces compressive strength and durability, limiting rubberised concrete use in structural applications [15]-[18]. Additionally, the soft and flexible nature of rubber particles can adversely impact

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the mechanical properties of the concrete, further complicating its use [19]. Another technical challenge involves the potential increase in the concrete's porosity when incorporating crumb rubber. The existence of rubber particles can generate voids within the concrete, impacting its density and potentially compromising its mechanical properties. This increased porosity may also affect the concrete's resistance to moisture ingress and chemical attacks, leading to reduced durability over time [1],[15],[19].

By addressing these challenges, nanomaterials have emerged as a promising solution [20]. Nanomaterials, owing to their minuscule dimensions and expansive surface area, can significantly enhance the properties of construction materials [21]-[22]. Nanomaterials can improve the interfacial bonding between rubber particles and the cement paste in rubberised concrete, consequently boosting the material's overall compressive strength and durability [23]-[24]. Various nanomaterials, including nano-silica, nano-clay, graphene oxide, and carbon nanotubes, have been explored for their potential to modify concrete's microstructure and mechanical properties [25]-[27].

The utilisation of nanomaterials in rubberised concrete can lead to several beneficial effects. For instance, nano-silica can fill the voids at the rubber-cement interface, creating a denser and more cohesive matrix [15],[28]-[29]. This results in improved load transfer and higher compressive strength. Similarly, carbon nanotubes can act as bridges across microcracks, enhancing the toughness and crack resistance of the concrete [30]-[31]. These enhancements mitigate the drawbacks associated with using crumb rubber and open up new possibilities for applying RC in structural and high-performance materials. Hence, through tailored nanomaterial formulations, it becomes possible to optimize the performance of rubberised concrete while overcoming technical hurdles and ensuring sustainable construction practices. The synergistic effects of nanomaterials and CR hold the potential to revolutionise the field of concrete technology, paving the way for advanced materials with superior properties and enhanced sustainability.

This review examines the latest research trends on nanomaterials' influence on rubberised concrete's compressive strength. This review synthesis the recent studies to enhance understanding of how nanomaterials can address the challenges posed by crumb rubber in concrete. The scope of the review encompasses an analysis of various types of nanomaterials used in very recent research, their mechanisms of action, and the resulting improvements in the mechanical properties of rubberised concrete. Additionally, the review will highlight the most promising nanomaterials for improving the performance of rubberised concrete, providing insights for future research and practical applications.

NANOMATERIALS IN RC

Nanomaterials are materials with structural features at the nanoscale, typically less than 100 nanometers in at least one dimension. At this scale, materials often exhibit unique physical, chemical, and biological properties that differ significantly from their bulk counterparts. These distinctive properties arise due to the high surface area-to-volume ratio, quantum effects, and the dominance of surface atoms, leading to enhanced reactivity, strength, and electrical and thermal conductivity, among other characteristics [32].

Concrete is a nanostructured, multi-phase composite material that evolves. It comprises an amorphous phase, crystals ranging from nanometer to micrometre sizes, and bound water. The properties of concrete and its degradation mechanisms span multiple length scales, from nano to micro to macro. Each scale's properties are influenced by those of the smaller scale beneath it. The amorphous phase, known as calcium–silicate–hydrate (C–S–H), acts as the "glue" that binds concrete together and is itself a nanomaterial [27].

Hence, nanomaterials can improve concrete's structural composition and mechanical characteristics by enhancing the hydration process, void filling, and bonding between aggregates and the cement matrix. These improvements can lead to increased compressive strength, reduced permeability, and enhanced durability. For that reason, many studies have used different types of nanomaterials to improve the performance of rubberised concrete. This review focuses on the most recent of those studies with the view to highlighting the current trend in the field of nanotechnology application in rubberised concrete. Table 1 summarises the most common nanomaterials used in rubberised concrete.

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Table 1 Summary of types of nanomaterials used in rubberised concrete

Type of Nanomaterial	Shape/ Morphology	Quantity (% wt. of cement)	Effect
Graphene nanoplatelets (GNPs) [33]	2D	0.1 to 0.7	0.4% GNPs increased compressive strength by 47.7%
Graphene oxide (GO) [34]	2D	0.01, 0.03, 0.05, and 0.1	Increments of 0.72%, 16.23%, 8.82%, and 6.48% were reported in the density, compressive strength, tensile strength, and flexural strength of RC after the inclusion of GO. GO reduced workability by up to 13.04% and air content by 1.5%.
Nanosilica (NS) [28]	0D	1.0	30.3% increase in early compressive strength
Carbon nanotubes (CNTs) [35]	1D	0.05 to 0.2	t a 0.1% CNT content, cement hydration increased by 8%, and the humidity diffusion coefficient decreased by 41%.
Nano Iron oxide (NIO) [36]	0D		NIO increased compressive strength by 24.82%, tensile strength by 6.58%, and flexural strength by 15.45% at 28 days
Nano Titanium dioxide (TiO_2) [23]	0D	1 to 2	At 28 days, rubberised concrete with 1.5% TiO_2 and 10% CR exhibited a compressive strength of 51.40 MPa, a tensile strength of 4.47 MPa, a flexural strength of 5.91 MPa, and an elastic modulus of 40.15 GPa.

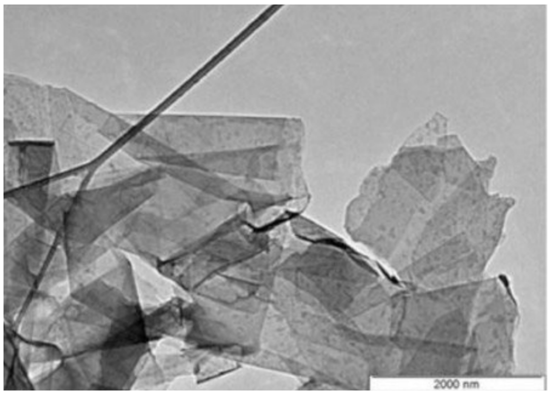
Graphene Nanoplatelets and Graphene Oxide in RC

Graphene Nanoplatelets (GnPs) are two-dimensional carbon nanomaterials consisting of multiple layers of graphene sheets, as shown in the TEM and SEM images in Figure 1. Due to their unique structure, these nanoplatelets exhibit exceptional mechanical, electrical, and thermal properties. With a high aspect ratio, large surface area, and excellent conductivity, GnPs have garnered significant interest in various industries, including electronics, aerospace, energy storage, and composites [37]-[38]. This nanomaterial has recently been used in rubberised concrete research.

Algaifi et al. [33] investigated rubberised concrete's acoustic and mechanical characteristics reinforced with GnPs. The research demonstrated decreased mechanical strength as the CR content increased. However, adding GnPs significantly reversed this trend, particularly at moderate levels of CR replacement and GnPs, resulting in a compressive strength more significant than that of the control sample with 0% CR replacement, as illustrated in Figure 2. For instance, when 0.4% GnPs were added, the concrete with 4% rubber achieved a compressive strength of 55.1 MPa, while the concrete with 11% rubber reached 49.5 MPa. Both strengths surpassed the control concrete's strength of 45.5 MPa.

Incorporating 0.4% GnPs alongside slag resulted in a notable enhancement in the compressive strength of

(a)



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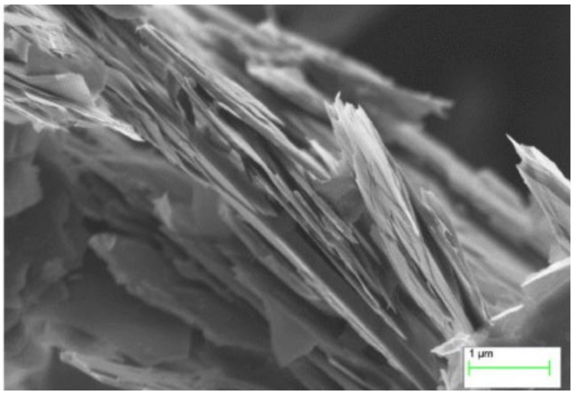

Figure 1 (a) TEM image of GnPs (b) SEM image of GnPs [39]

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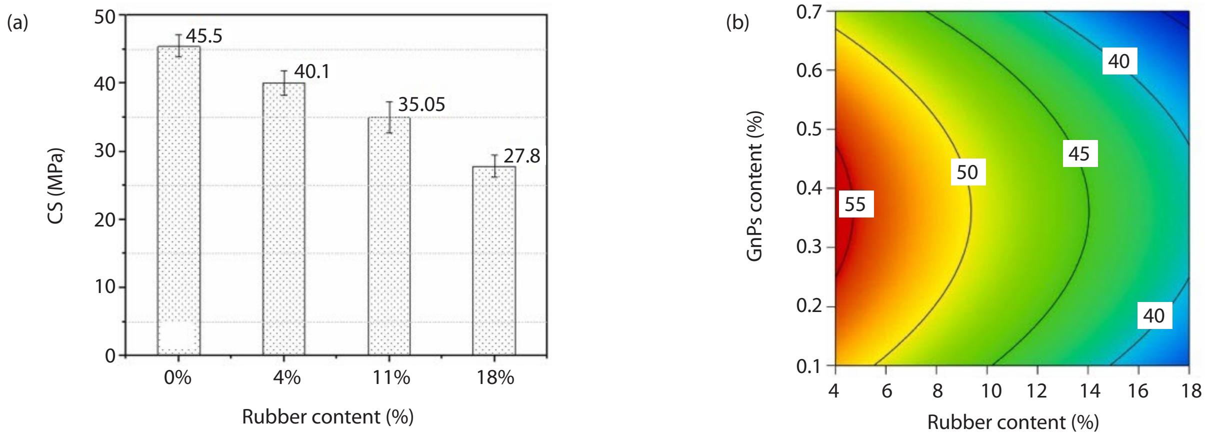

Figure 2 Influence of CR and GnPs on the compressive strength of rubberised concrete [33]

rubberised concrete by 47.7%, rising from 27.85 MPa to 41.1 MPa. This value closely approached that of the control specimen at 45.5 MPa. This improvement is ascribed to the collaborative influence of GnPs and slag, which fortify the concrete matrix at both micro and nano scales. GnPs, boasting a tensile strength of up to 130 GPa, adeptly absorb loads and redistribute stresses within the cement matrix, refining the concrete microstructure when subjected to load. Moreover, GnPs act as effective fillers, forming a dense barrier against crack propagation, thereby augmenting the overall performance of the concrete [40]. Additionally, the distinctive characteristics of slag, including its particle size distribution and expansive surface area, expedite a swift chemical reaction between calcium hydroxide ($Ca(OH)_2$) derived from cement hydration and the silicates in slag. This reaction fosters the generation of supplementary C-S-H gels, further enhancing the concrete's mechanical properties [33].

Graphene oxide (GO) has garnered significant attention from researchers due to its exceptional properties, surpassing those of other carbon-based nanomaterials [41]-[43]. Despite its potential, the application of GO in rubberised concrete has been relatively unexplored, with only a limited number of studies available. Chaturvedy et al. [34],[44] have published two key studies investigating this innovative approach, highlighting the promising results and potential benefits of incorporating GO into rubberised concrete. Using different optimisation methods, the influence of GO on the properties of high-strength RC was assessed [34].

The experimental results show that CR significantly reduces compressive strength, which can be mitigated by adding GO. Adding GO with constant CR contents of 5%, 10%, and 15% improved compressive strength by 11.95%, 10.8%, and 16.23%, respectively. Similarly, a 13.45% increase in the compressive strength of RC was observed in another study [44]. These strength enhancements are due to GO improving C-S-H formation and acting as filler that occupies pores and encourages a more compact structure. The surface area of GO acts as a site for the initiation of hydration products, along with its oxygen functional groups interacting with cement chemicals to form strength-enhancing crystals [43].

Maximum compressive strength was achieved with 0.05% GO content. Beyond this, at 0.1% GO, compressive strength decreased due to dispersion issues and nanoparticle agglomeration. Well-dispersed GO sheets exhibit greater efficacy in enhancing mechanical properties than agglomerated sheets. They leverage their larger surface area to enhance cement hydration [45]-[46].

Nanosilica (in Rubberised Concrete

Nanosilica (NS), also known as silica nanoparticles, refers to very fine particles of silicon dioxide (SiO_2) with dimensions typically in the nanometer scale [29], as shown in powdered form in Figure 3. These nanoparticles possess distinctive characteristics attributable to their minute size, notably a high surface area, enhanced reactivity, and improved mechanical properties when incorporated into various materials [47]. On the other hand, colloidal nano-silica refers to NS particles that are dispersed in a liquid medium, typically

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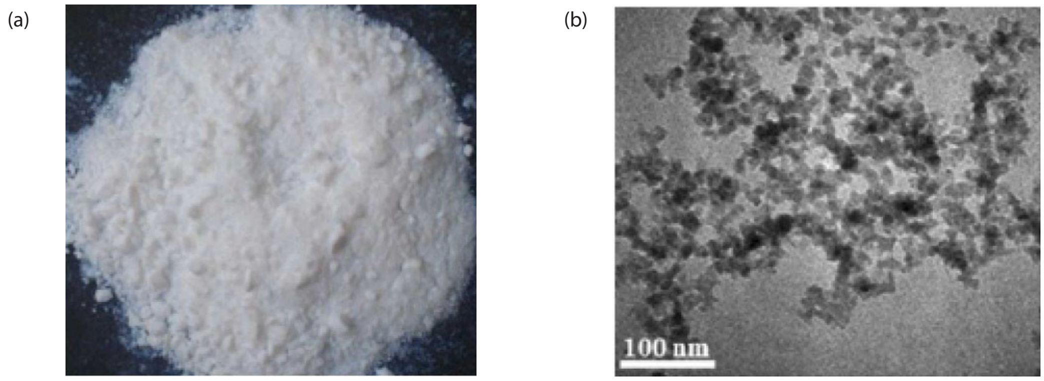

Figure 3 (a) Nano silica Powder (b) TEM image of Nano silica [49]

water. These colloidal suspensions contain nanoscale silica particles stabilized to prevent aggregation or settling [48].

NS in concrete has garnered significant attention due to its capacity to enhance strength. Some current research is on its use in rubberised concrete. Xu et al. studied the influence of colloidal NS (CNS) and its enhancement mechanism on the strength of RC [28]. Modifying rubberised concrete with CNS significantly enhances its strength, as shown in Figure 4. The rubber particles and cement mortar exhibit a strong bond in rubberised concrete modified with CNS. Element distribution analysis indicates a reduction in the thickness of the ITZ, and CNS incorporation effectively improves the ITZ's microstructure. The mechanism by which CNS enhances strength and ITZ microstructure involves three aspects [28]:

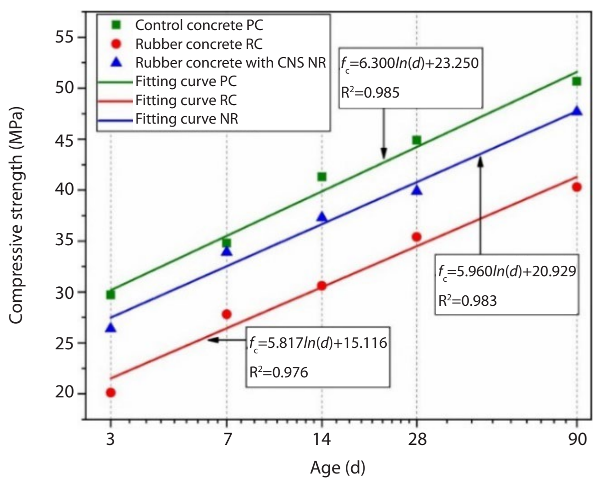

Figure 4 Comparison between normal concrete, normal rubberised concrete, and rubberised concrete containing NS [28]

1. NS particles adsorb onto the rubber surface, acting as nuclei for hydration reaction, strengthening the adhesion between the cement matrix and rubber.
2. The substantial pozzolanic reactivity of NS greatly facilitates the reaction between NS and calcium hydroxide (CH), forming C-S-H.
3. The presence of NS enhances the compactness of the ITZ microstructure through its filling effect.

As shown in Figure 5, the mechanism of the ITZ between CNS-modified rubber and cement has two primary effects. Firstly, due to their high surface energy, NS particles exhibit a strong adsorption force and effectively adhere to the CR surface within the rubber-paste ITZ. Secondly, the considerable pozzolanic reactivity of NS greatly enhances the reaction between NS and CH to form C-S-H. In forming the ITZ between CNS-modified rubber and mortar, NS particles adhered to the rubber surface function as nuclei for cement hydration. This core effect facilitates the effective combination of cement hydration products and rubber particles, improving the initial defects of the rubber-cement ITZ. The significant pozzolanic reactivity of NS decreases the presence of CH particles. The resulting C-S-H makes the ITZ microstructure more compact. Additionally, NS particles occupy the micro- and nanopores within the cement slurry, increasing the particle size gradient at the nanometer scale, which enhances the density of the concrete microstructure [50].

Furthermore, Adamu et al. [51] recently conducted a study on utilising artificial neural networks (ANN) to predict the mechanical properties of RC containing NS and fly ash. Consistent with the findings of previous

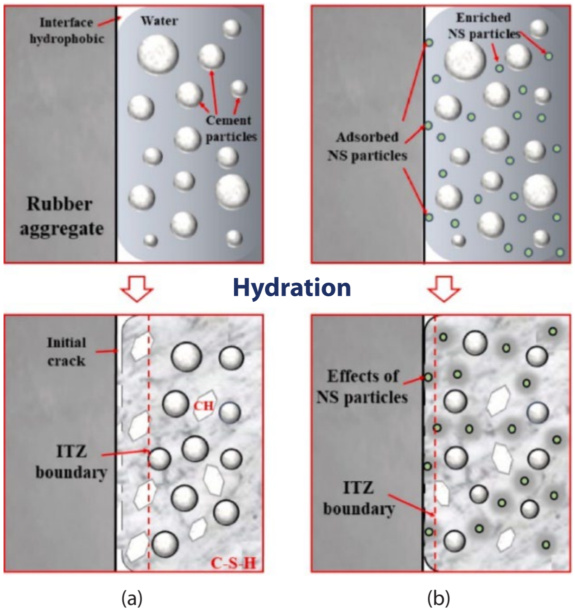

Figure 5 CNS mechanism of ITZ modification (a) normal concrete (b) CNS concrete [28]

studies, incorporating CR diminishes the mechanical properties of concrete but enhances other attributes. NS is used to improve the concrete and counteract the detrimental effects of CR. The ANN model, trained and tested with experimental data, exhibited high prediction accuracy for the mechanical properties, with minimal proportional deviation mean (MoD) values close to zero. The results emphasize that NS significantly bolsters the mechanical properties of rubberised concrete, suggesting that it is a suitable material for sustainable construction. The model's mean square error (MSE) and coefficient of determination (R^2) values further corroborate the reliability of the predictions.

Carbon Nanotubes in Rubberised Concrete

Carbon nanotubes (CNTs) are cylindrical structures composed of carbon atoms arranged in a hexagonal pattern. They can be single-walled (consisting of a single layer of carbon atoms) or multi-walled (multiple layers nested concentrically), as shown in Figures 6a and 6b, respectively. Carbon nanotubes exhibit exceptional mechanical strength, high conductivity, and unique thermal properties due to their atomic structure. These nanotubes can be metallic or semiconducting based on their structure, offering versatility in various applications [52]-[53].

Despite its popularity due to exceptional properties such as high strength and stiffness, lightweight, high aspect ratio, electrical conductivity, and thermal conductivity, recent research on using CNTs in rubberised concrete is limited. The most recent study involving CNTs in RC was conducted by Gao et al. [35], who analyzed and modelled the mechanism of CNTs in regulating the internal curing of rubberised concrete. The research demonstrated that due to the microscopic effect of the CNTs, as shown in Figure 7, they effectively regulated the humidity of RC, establishing a dual-phase model

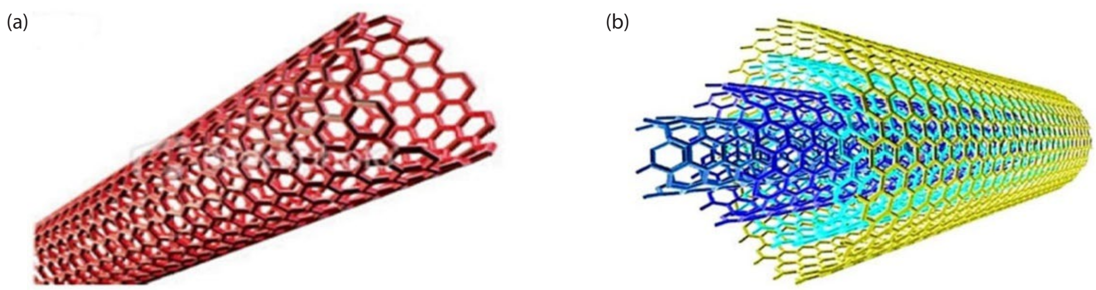

Figure 6 (a) Single-walled CNT (b) Multi-walled CNT [53]

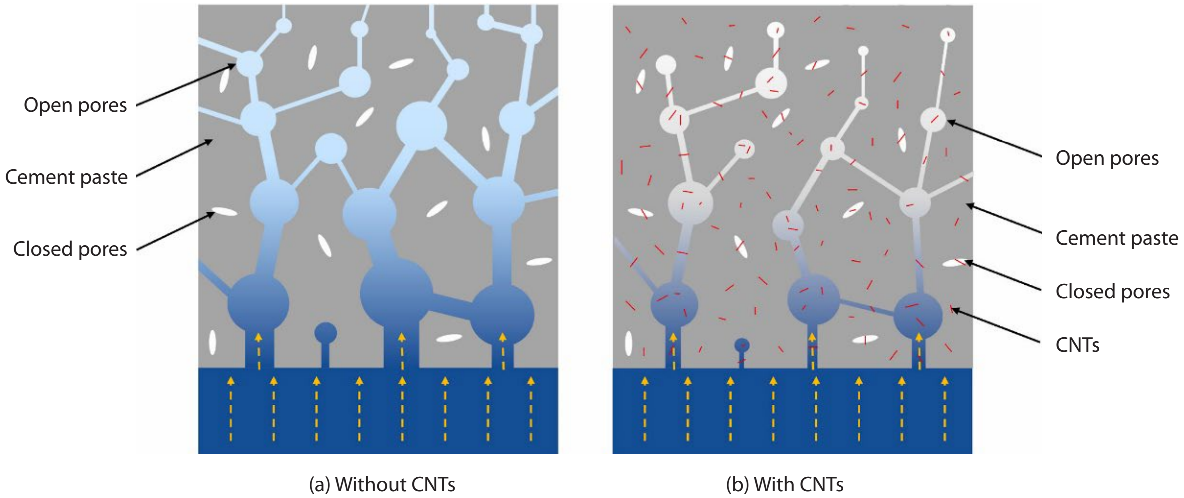

Figure 7 Schematics of the pore-water interaction inside a concrete (a) without CNTs (b) containing CNTs [35]

integrating cement hydration and moisture diffusion to simulate the moisture dynamics of CNTs modified rubberised concrete. Molecular dynamics simulations demonstrated that van der Waals forces exerted by CNTs induced a vacuum layer, expediting hydration kinetics, resulting in an 8% increase in cement hydration and a 41% reduction in moisture diffusion coefficient with 0.1% CNTs inclusion. The proposed moisture model accurately forecasted moisture dispersion across varied conditions, advocating for a 0.1% CNTs dosage to achieve an optimal equilibrium between mechanical strength and shrinkage performance in rubberised concrete [35].

Furthermore, Shou et al. [30] studied the durability and performed a fractal analysis of CNTs-modified rubberised concrete. The study explored the impact of CNTs in rubberised concrete, focusing on durability and pore structure. Different CNTs concentrations were added to RC with varying rubber content. Results revealed that CNTs and rubber particles notably decreased water absorption and chloride penetration compared to standard concrete. Rubber addition increased coarse capillary and macropores, with CNTs mitigating pore structure deterioration. The study highlighted scale-dependent fractal characteristics, showing CNTs' influence on pore coarsening. Notably, 0.1% of CNTs enhanced pore structure, permeability, and residual strength in RC, indicating the beneficial role of CNTs in improving the properties of the RC.

Nano Iron Oxide in Rubberised Concrete

Another important nanomaterial that has been investigated in rubberised concrete recently is Nano Iron Oxide (NIO), which refers to iron oxide particles that are at the nanometer scale, typically less than 100 nanometers in size. The particles exhibit unique properties at this size, including high surface area, magnetic properties, chemical reactivity, mechanical strength, and optical properties [54]. A recent study by Saini et al. [36] assessed the effect of NIO on the properties of RC. It was observed that while CR decreased workability and density by up to 13.46% and 5.76%, respectively, and reduced compressive, tensile, and flexural strengths by 20.69%, 22.49%, and 24.16%, respectively, the addition of NIO significantly mitigated these adverse effects. With NIO, compressive strength increased by 24.82%, tensile strength by 6.58%, and flexural strength by 15.45% at 28 days. Furthermore, a life cycle assessment indicated reduced CO_2 emissions with minimal increase in embodied energy, highlighting NIO's role in enhancing RC properties and promoting sustainable construction practices.

Nano Titanium Dioxide in RC

Titanium dioxide (TiO_2) nanoparticles, known as Titania, are increasingly used in cementitious composites due to their superior photochemical properties, stability, and cost-effectiveness. These nanoparticles can be utilised either in powdered form or as a solution, as illustrated in Figures 8a and 8b, respectively. These nanoparticles accelerate the hydration process by filling microscale voids and creating nucleation sites for the early formation of hydration products. Research has

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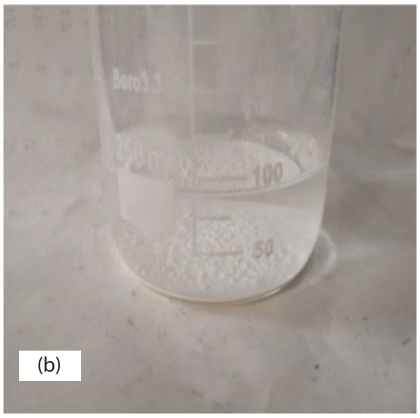

Figure 8 (a) Nano TiO_2 powder (b) Solution of TiO_2 [56]

demonstrated significant advantages of incorporating TiO_2 into cement matrices, highlighting their potential to enhance the material's overall performance [55].

Abdullah et al. [23] conducted one of the most recent studies on the use of TiO_2 in rubberised concrete, where the effect of the nanomaterial on the mechanical and durability properties was assessed. The integration of TiO_2 nanoparticles profoundly influences the compressive strength (CS) of rubberised concrete. As presented in Figure 9, adding TiO_2 enhances the CS of various RC mixtures after 28 days. A significant peak in CS, reaching 51.40 MPa, was attained with a mix containing 10% CR and 1.5% TiO_2 relative to the cement's weight [23]. Conversely, the lowest CS of 38 MPa was registered with 30% CR and 2% TiO_2. The strength diminishes as CR content escalates due to its inadequate adhesion with the cement matrix and its hydrophobic nature, impeding water interaction and bonding [57]-[59].

Including TiO_2, particularly up to 1.5%, fortifies the RC matrix by enhancing the hydration process. TiO_2 engages in the pozzolanic reaction, fostering the generation of C-S-H, which is pivotal for concrete strength [23]. However, heightened TiO_2 concentrations deteriorate performance due to uneven nanoparticle dispersion and excessive water absorption, constraining C-S-H gel formation. Optimal TiO_2 integration (1.5%) effectively bolsters CS by 22.6% at 28 days, underscoring its potential to counteract CR's adverse effects and enhance rubberised concrete's mechanical properties. This aligns with prior research indicating TiO_2's efficacy in reinforcing concrete microstructure as an effective nanofiller [23].

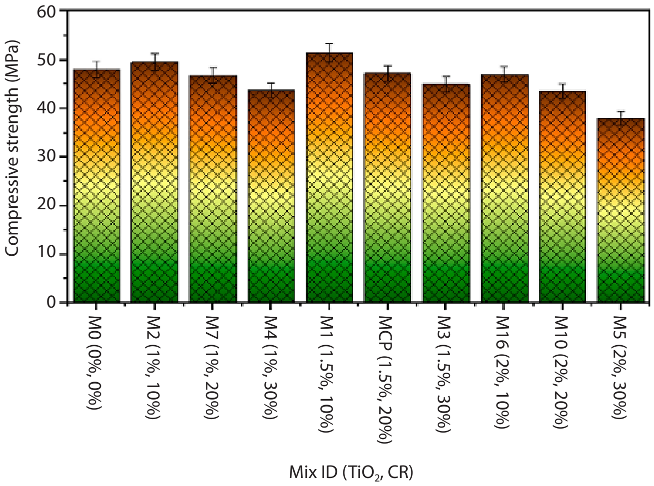

Figure 9 Compressive strength of RC containing TiO_2 [23]

Furthermore, to enhance concrete's performance and photocatalytic properties, CR modification using Tannic acid (TA) and TiO_2 was done by Zhu et al. [60]. Figure 6 illustrates that the concrete with CR modified by TA and TiO_2 exhibits early compressive strength that exceeds the reference concretes. This improvement is due to the modified CR's better compatibility with the mortar and its "hard shell" surface, alleviating the negative effects typically associated with CR. During hydration, the mortar adheres and bridges to the modified CR surface, filling voids and creating a more tightly bound internal structure. As a result, this concrete shows increased compressive strength compared to the reference mix.

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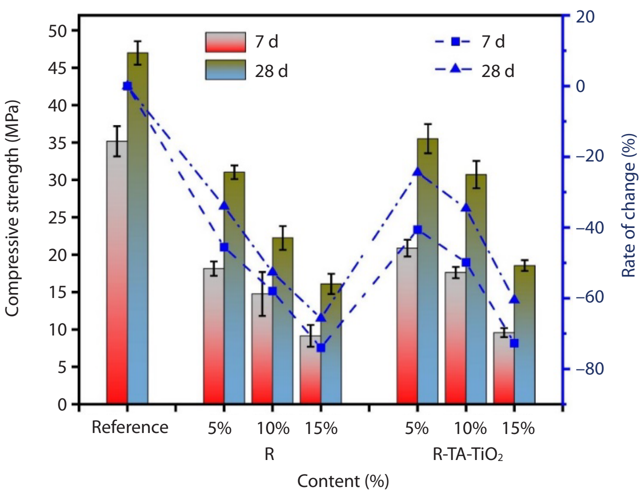

Figure 10 Compressive strength of reference concrete, rubberised concrete, and rubberised concrete containing TA and TiO_2 modified CR [60]

CONCLUSION

This paper reviewed recent developments in the use of nanomaterials in rubberised concrete, focusing on studies from 2023 to 2024. Despite the promising enhancements brought by nanomaterials like graphene oxide (GO), graphene nanoplatelets (GNPs), nanosilica (NS), carbon nanotubes (CNTs), and titanium dioxide (TiO_2), their application in rubberised concrete is still relatively underexplored. The observed improvements in strength and the mitigation of the negative effects of crumb rubber (CR) are often linked to mechanisms such as pore filling, nucleation, and enhanced hydration reactions. Future research should focus on optimizing the type and dosage of these nanomaterials, conducting long-term durability studies, and performing cost-benefit analyses to ensure practical viability. Exploring emerging trends like bio-based and waste-based nanomaterials, alongside advanced characterization techniques, offers exciting opportunities for further enhancing rubberised concrete's performance and sustainability, ultimately contributing to waste tyre disposal solutions on a global scale.

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