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Advances in the durability of recycled aggregate concrete modified with nanosilica

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In this article, the latest achievements in the study of concrete modified with nanosilica with a secondary filler are systematically reviewed. The key aspects of durability are studied, including water absorption, chloride ion permeability, freeze-thaw resistance, and acid resistance. The mechanisms by which silica improves the durability of concrete with secondary filler are studied, providing valuable theoretical and technical ideas for its application and improvement.
Keywords: Nanosilica, recycled aggregate concrete, durability, modification mechanisms

Досягнення в довговічності переробленого бетону, модифікованого нанокремнеземом. *Fang Qin, Yuqing Yan*

В цій статті систематично розглядаються останні досягнення в дослідженні модифікованого нанокремнезему бетону з вторинним наповнювачем. Досліджені ключові аспекти довговічності, включаючи водопоглинання, проникнення хлорид-іонів, стійкість до замерзання-відтаювання та кислотостійкість. Вивчаються механізми, за допомогою яких кремнезем покращує довговічність бетону з вторинним заповнювачем, що надає цінні теоретичні та технічні ідеї для його застосування та вдосконалення.

1. Introduction

Recycled aggregate concrete (RAC) has become an important area of research and development of sustainable construction waste management technologies. This innovative building material not only reduces landfill waste but also conserves natural resources by decreasing the demand for sand and stone extraction. Such advancements align with national strategic goals for green, low-carbon, and sustainable development. However, the higher porosity of recycled aggregates compared to natural aggregates creates significant challenges that negatively affect the durability of RAC [1-4].

In recent years, nanomaterials have attracted attention due to their unique physicochemical properties, versatility, cost-effectiveness, and nucleation effects [5], making them valuable for enhancing the performance of cement-based materials [6, 7]. Nanosilica (NS) is a nano-material with pozzolanic characteristics [8, 9]; it can interact with calcium hydroxide (CH) in the interfacial transition zones (ITZs) of recycled aggregates, leading to the formation of calcium silicate hydrate (C-S-H) gel that fills the pores in old mortar, thereby improving the structure of the ITZs. Numerous studies indicate that the incorporation of NS enhances the strength of RAC at various stages, especially in the early stage of curing, with early strength increases ranging from 6.7% to 53% compared to control groups, while later strength variations range from 1.7% to 28% [10-12]. Although an appropriate dosage of NS can effectively enhance the compressive strength of RAC, excessive amounts may result in strength reduction or even negative growth [13, 14], probably due to the agglomeration of NS particles that inhib-

Table 1 Enhancement of recycled aggregate (RA) performance using NS after 28d.

<table>
<tr><th>Enhancement methods</th><th>NPs</th><th>Advantages</th><th>Disadvantages</th><th>Ref.</th></tr>
<tr><td rowspan="4">Direct incorporation</td><td rowspan="4">NS powder or solution</td><td rowspan="4">· Fill the pores in the RA and dense the microstructure;
· Improve the durability and mechanical properties of RAC;
· Increased functionality
· Volcanic ash effect forms more C-S-H gel</td><td rowspan="4">· Limited properties improvement
· Limited enhancement effect on ITZs</td><td>[16, 23-26]</td></tr>
<tr><td>[10, 12, 13, 16, 27-29]</td></tr>
<tr><td>[22, 30, 31]</td></tr>
<tr><td>[32-34]</td></tr>
<tr><td>Pre-soaking</td><td>NS solution</td><td rowspan="2">· Make ITZs dense and strong;
· Enhance the bonding between RA and the new cement paste;
· Improve the quality and strength of RA.</td><td rowspan="2">· Uniformity is difficult to control;
· Costs increase;
· Waste solution causes environmental problems</td><td>[17, 18, 20, 21, 31, 35, 36]</td></tr>
<tr><td>Pre-spraying</td><td>NS solution</td><td>[17, 18]</td></tr>
</table>

it the hydration process of cement [15, 16]. Consequently, the optimal dosage of NS is typically recommended to be between 2% and 3%. Furthermore, research indicates that the spraying method is the most effective for improving RAC performance, followed by the soaking method, while direct incorporation yields relatively less favorable results [17, 18].

In addition to strength enhancement, the durability of RAC is critical, as the service life of a structure largely depends on the durability of its cement-based materials. As a result, the role of NS in enhancing the durability of RAC has attracted considerable interest. Research has shown that the inclusion of NS substantially improves various durability indicators, including water absorption, chloride ion permeability, corrosion resistance, acid resistance, and high-temperature resistance. This paper reviews the current research on the durability of NS-modified recycled coarse aggregate concrete, providing a theoretical foundation and technical support for further enhancements in the durability of RAC.

2.Nanosilica modification approaches

NS typically ranges from 8 to 100 nanometers in size and it appears as an amorphous white powder or transparent solution composed of SiO_2 molecules with an amorphous structure. Within this structure, SiO_2 tetrahedra are arranged in a short-range ordered manner through corner-sharing. The smaller size of silicon ions relative to oxygen ions enables them to fill the voids within the tetrahedra, resulting in a unique amorphous configuration. This three-dimensional chain-like structure provides NS with a high number of unsaturated bond sites and hydroxyl groups, which contribute to its exceptional reactivity and favorable surface chemical properties [11]. Considering its high specific surface area, excellent dispersibility, stability, and enhanced pozzolanic activity [19], the application of NS in RAC has been the subject of extensive investigation.

Common methods for enhancing the performance of recycled aggregates (RA) primarily involve two approaches: modifying the RA itself and increasing the density of the RAC matrix. Table 1 summarizes various widely employed methods for improving RA performance. The three principal methods for NS modification of RAC include spraying, soaking, and direct incorporation (either singly or in combination). Existing studies demonstrate that NS significantly enhances the properties of RA. For instance, treatment of RA with NS particles via soaking resulted in a 21.4% reduction in water absorption and a 29.3% decrease in specific gravity [20]. Additionally, research by Zeng et al. [21] revealed that after a 24-hour immersion in a NS suspension, the surface microhardness of RA increased from 42.82 HV to 54.12 HV. This enhancement is attributed to the ability of NS to penetrate the pores of the old mortar, thereby significantly increasing the density of the RA surface [22].

3 Durability

3.1 Water Absorption

The incorporation of NS into RAC has been shown to significantly reduce its water absorption. Research conducted by Erdem et al. [37] indicates that RAC samples containing 0.5% to 2% NS exhibit reductions in water permeability ranging from 28% to 84% compared to control specimens. Younis et al. [38] and Hosseini et al. [1] further demonstrated that appropriate NS dosages markedly enhance the water absorption performance of RAC, with tests assessing absorption over periods of 24 hours and 0.5 hours, respectively. Additionally, Mukharjee and Barai [39] reported that replacing natural coarse aggregate (NCA) entirely with RA resulted in an increase in water absorption from 4.74% to 6.60%. However, the inclusion of NS reduced the absorption rate to 4.58%, which is consistent with the results of earlier studies [13, 30].

In comparison to direct incorporation methods, the soaking technique is notably more effective, yielding a further 15% reduction in RAC water absorption [31]. This improvement is attributed to the deposition of NS particles on the surface of the RA, which enhances the microstructure of the ITZs by repairing existing cracks and porous areas [40]. Supporting this, Singh et al. [20] and Li et al. [17] found that pre-treatment of RAC with large-diameter NS led to reductions in initial and secondary water absorption rates by 35.9% and 38.4%, respectively, achieving results comparable to those of NAC. The effectiveness of NS in this context is primarily due to its ability to fill pores and cracks within the recycled coarse aggregate, thereby increasing resistance to moisture penetration [41]. Furthermore, the high reactivity of NS facilitates the formation of additional C-S-H gel, which fills voids in the concrete matrix and effectively diminishes the water absorption of RAC by lowering porosity and increasing density [42].

In geopolymers that incorporate recycled fine aggregates, the addition of NS also contributes to a reduction in water absorption. However, this positive effect diminishes as the NS content increases [43]. This phenomenon can be attributed to a by the decrease in NS dispersion, which enhances agglomeration and leads to the formation of weak zones within the ITZs; this in turn, leads to larger pores that facilitate moisture penetration [44]. Research by Agarwal et al. [45] corroborated these findings, showing that when the NS content reached 5%, the water absorption of RAC significantly increased. This suggests that excessive NS dosage may induce microstructural degradation, thereby undermining the overall performance of the material.

3.2 Chloride ion penetration

Numerous studies have demonstrated the significant positive effects of NS on the resistance of RAC to chloride ion penetration. Shaikh et al. [30, 31] reported that incorporating 2% NS reduced the chloride ion penetration depth of RAC specimens by 28% to 31% after 28 days. Similar findings were reported by Sahu et al. [46]. Ying et al. [27] conducted rapid chloride migration tests to compare the effects of NS and nano-titanium (NT) on the resistance of RAC to chloride ion penetration. Their results indicated that the diffusion coefficient ranged from 0.6 to 1.2 × $10^{?12}$ m²/s, with NT providing superior performance compared to NS. Some researchers suggest that NS enhances resistance to chloride ions by refining the pore structure of RAC [47, 48], resulting in a denser microstructure [49-51].

In comparison to the direct mixing method, RAC specimens treated with NS by impregnation exhibited substantial improvements in both resistance to chloride ion penetration and electrical charge transport, outperforming even NAC [17]. Impregnation of RA with a slurry composed of NS and nano-calcium (NCa) significantly increased the resistance of RAC to chloride ion migration, an effect attributed to the enhanced ITZs [52]. Shaikh et al. [31] compared the effects of internal mixing and pre-immersion methods on the resistance of RAC to chloride ion penetration and found that the pre-immersion method gave superior results. Li et al. [53] also showed that prespraying RA with NS suspension enhanced the resistance of RAC to chloride ion penetration, while no significant differences were observed between samples with different NS contents. When NS suspension was sprayed onto the RA surface, a dense layer formed, characterized by a low calcium-to-silicon ratio (C/S < 1.0) in the C-S-H gel. This “aggregate surrounding enhancement” effect, similar to “aggregate coating,” redirected the chloride ion penetration path from passing through the aggregate to bypassing the aggregate edges, significantly improving the resistance of RAC to chloride ion penetration [54].

Furthermore, Alhawat et al. [10] explored the effects of NS on the corrosion resistance of steel reinforcement in RAC exposed to chloride ion environments. Their results demonstrated that the corrosion rate of 100% RAC specimens containing 1.5% NS was reduced by 40% to 52% over 2 to 15 days, compared to specimens without NS. With an increase in the NS content to 3%, the corrosion rate decreased by another 5–25%. Zeng et al. [21] also observed that RA impregnated with NS significantly improved the corrosion resistance of rebar in RAC. The corrosion current density of rebar in unmodified RAC (CRAC) was higher than that in modified RAC (MRAC), and rebar corrosion in CRAC began approximately four weeks earlier than in MRAC. Corrosion-induced cracks on the sides of CRAC specimens appeared two weeks earlier and were more extensive than those on MRAC specimens. This accelerated corrosion process in CRAC could be attributed to earlier chloride ion migration to the rebar surface under the influence of direct current, whereas MRAC exhibited better protection due to NS-induced modifications.

However, a few studies have reported adverse effects from excessive NS incorporation [55]. Agarwal et al. [45] found that when the NS content was too high, the chloride ion permeability of RAC specimens containing 60% RA increased. Similarly, Nuaklong et al. [43] observed that as the NS content increased from 0% to 3%, the chloride penetration depth in geopolymer RAC after 90 days increased by 96.5% to 191%. This deterioration in chloride ion resistance may be associated with a reduced CaO/SiO_2 ratio in the C-S-H gel, which reduces the permeability resistance of RAC.

3.3 Freeze Resistance

Numerous studies have demonstrated that the freeze resistance of RAC is significantly lower than that of conventional concrete. However, the incorporation of NS has been shown to substantially enhance the freeze resistance of RAC. Liu [56] reported that with an increasing number of freeze-thaw cycles, both the compressive strength and the relative elastic modulus of recycled concrete decreased progressively, initially showing a decrease in the rate of mass loss followed by an increase. The optimal dosage of NS for improving compressive strength, relative elastic modulus, and reducing mass loss rate was identified as 4%. Notably, within the same number of freeze-thaw cycles, freeze resistance initially increased at higher NS content and then decreased.

The internal compactness of concrete deteriorates following freeze-thaw cycles, leading to ongoing freeze-thaw damage as the number of cycles accumulates [57]. While a reasonable amount of NS can enhance the freeze resistance of RAC, excessive NS content may result in agglomeration, which increases the number of internal defects and negatively affects freeze resistance. As a result, the freeze resistance of RAC increases up to a certain level of NS content and then begins to decrease. This behavior can be attributed primarily to the high pozzolanic activity and filler effects of NS, which contribute to the densification of concrete and its resistance to freeze-thaw damage. However, an excessive amount of NS can lead to an increase in unhydrated NS particles, which will result in a decrease in the specific surface area and thus a decrease in the effectiveness of NS in enhancing freeze resistance. Therefore, it is essential to determine the optimal dosage of NS for incorporation into RAC formulations. Wang et al. [58] identified that the attached mortar on the surface of recycled aggregate (RA) plays a crucial role in the frost resistance of RAC. During freeze-thaw cycles, microcracks in the concrete initially concentrate at the old mortar interface, leading to the disintegration of the RAC, which subsequently propagates to the new mortar and results in severe freeze-thaw damage to the concrete. Therefore, it is recommended to utilize NS solutions for pre-soaking and pre-spraying treatments of RA to enhance the frost resistance of RAC. Gokce et al. also reached a similar conclusion [59].

Furthermore, by using 1.5% NS emulsion and 1.5% polyvinyl alcohol (PVA) to modify RA in combination with replacing 10% cement with fly ash (FA), the freeze-thaw resistance of RAC under mechanical loading and saline solution exposure was significantly improved. Specifically, the compressive strength loss of the samples was reduced from 19.8% to 9%, 10.3%, and 7.5%, respectively.

3.4 Acid Resistance

Sahu et al. [46] investigated the acid resistance of recycled fine aggregate concrete (RFAM) containing 10% to 70% RA. Their findings revealed that as the concentration of sulfuric acid (H_2SO_4) increased, the compressive strength of the samples after 28 days gradually declined. This reduction can be attributed to the chemical reactions occurring between sulfuric acid and the hydration products of cement, specifically CH and calcium silicate hydrate (C-S-H), which form gypsum. The gypsum further reacts with tricalcium aluminate (C-A) to produce ettringite [60], as illustrated in the following reactions [61]:

$$Ca(OH)_2 + H_2SO_4 \rightarrow CaSO_4 \cdot 2H_2O \quad (1)$$

$$CaO \cdot SiO_2 \cdot 2H_2O + H_2SO_4 \rightarrow CaSO_4 + Si(OH)_4 + H_2O \quad (2)$$

$$3CaO \cdot Al_2O_3 \cdot 12H_2O + 3CaSO_4 \cdot 2H_2O + 14H_2O \rightarrow 3CaO \cdot Al_2O_3 \cdot 3CaSO_4 \cdot 32H_2O \quad (3)$$

The incorporation of NS has been shown to partially mitigate the strength loss associated with acid attack in RFAM. Notably, this compensatory effect becomes increasingly pronounced with rising acid concentrations.

Conversely, Rao et al. [62] found that in 3% and 5% hydrochloric acid or sulfuric acid solutions, increasing the NS content from 2% to 3% resulted in decreased acid resistance of the RAC samples. Furthermore, Nuaklong et al. [43] showed that the addition of NS in geopolymers diminished the sulfate resistance of RAC. This deterioration in resistance may be caused by unreacted NS filling large voids, thereby reducing the available space for expansive reactions, leading to increased internal pressure and subsequent structural failure of the RAC [63, 64].

3.5 Other Durability Aspects

Studies examining additional aspects of the durability of NS-containing RACs are limited. Carbonation tests indicate that the incorporation of a small amount of NS can reduce the carbonation of cement paste, especially affecting C-S-H [65]. Sahu et al. [46] noted an improvement in the carbonation resistance of RFAM with the addition of NS. However, the effect of NS on the carbonation resistance of RAC remains a topic of debate [66]. Although most researchers claim that NS enhances the carbonation resistance of RAC [60, 67], some studies suggest that it may have adverse effects.

Yonggui et al. [68] reported that at elevated temperatures ranging from 200°C to 400°C, the compressive strength of 100% RAC improved with the inclusion of NS; however, at 600°C, a declining trend in compressive strength was observed, with residual strength consistently decreasing. Wang et al. [69] investigated the modification of recycled concrete by simultaneously adding both NS and basalt fiber (BF). The results demonstrated that the combined use of NS and BF effectively enhanced the high-temperature performance of recycled concrete. When BF was added independently at the same temperature, both the width and number of cracks decreased as BF content increased, primarily due to the bridging effect of BF that limits crack propagation. Additionally, it was concluded that, under consistent conditions, the quality of BF did not exhibit a systematic influence on high-temperature compressive strength [70]. However, with increasing mass fractions of NS, the amount of C-S-H gel per unit volume of mortar also increased, leading to a gradual loosening of the structure and varying degrees of reduction in compressive strength.

4. Conclusions

NS significantly enhances the durability of RAC through mechanisms such as nucleation effects, filling action, and pozzolanic activity. The incorporation of nanoscale particles effectively fills voids within the concrete matrix, improving density and the structure of the ITZs, and promoting the formation of C-S-H gel. These enhancements collectively contribute to improved overall performance. Furthermore, the durability improvement achieved by using NS in RAC is evident in its microstructural reinforcement mechanisms, notably enhancing resistance to chloride ion intrusion and water absorption. Despite extensive research on RAC, several aspects warrant further investigation to facilitate broader applications:

(1) *Carbonation and Freeze-Thaw Resistance*: Significant progress has been made in understanding the carbonation and freeze-thaw performance of recycled concrete, particularly through qualitative analyses using microhardness tests and scanning electron microscopy. However, studies of microstructural evolution during carbonation are limited, and a comprehensive predictive model for carbonation depth in RAC remains undeveloped. Additionally, existing freeze-thaw studies predominantly focus on singular conditions, neglecting the coupled effects of corrosive ions on the freeze-thaw performance of RAC. There are also no systematic studies on the freeze-thaw behavior of fibre-reinforced RAC under challenging conditions. Future research should prioritize the durability of fiber-reinforced concrete under freeze-thaw conditions, particularly exploring perfor-

mance degradation and damage mechanisms influenced by multiple factors. Furthermore, a deeper investigation into early warning mechanisms for freeze-thaw damage in practical engineering applications is essential to provide reliable technical support for the long-term service of RAC.

(2) *Resistance to Salts*: While numerous studies have examined the diffusion behaviors of chloride (Cl^-) and sulfate (SO_4^{2-}) ions in recycled concrete, the micro-level mechanisms of corrosion deterioration remain insufficiently elucidated. Future research should focus on analyzing durability damage in multi-ion environments (e.g., SO_4^{2-}, Cl^-, Mg^{2+}), especially under the coupled effects of complex environmental conditions such as freeze-thaw cycles and wet-dry cycles. Developing a diffusion database for chloride and sulfate ions in practical engineering contexts is crucial for optimizing material design and application.

(3) *Mechanisms of Performance Enhancement*: Although the mechanisms by which NS enhances the performance of recycled coarse aggregate concrete have been widely studied, existing studies mainly focus on individual factors of degradation mechanisms, with a lack of investigation into the effects of multi-field coupling under complex service conditions. Long-term performance monitoring data for RAC is scarce, and the comprehensive influences of loading, salt attack, and freeze-thaw cycles in actual concrete service environments complicate the understanding of these mechanisms. Systematic research using multi-field coupling models is urgently needed. Future studies should focus on exploring the performance degradation mechanisms of NS-modified RAC under multidimensional and multi-interface conditions while developing more reliable damage assessment metrics and early warning systems. Furthermore, methods for assessing the durability of recycled coarse aggregate concrete based on microstructural control need further refinement because the lack of theoretical support creates significant problems in practical engineering application. Consequently, future research should strengthen the integration of laboratory studies with real-world applications, providing a more scientific and comprehensive theoretical and practical foundation for the sustainable application of RAC.

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