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Effects of nano-silica on mechanical properties of recycled concrete: a review

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This article provides a literature review on the use of nanosilica to improve the surface of recycled nanosilica aggregate and modify the properties of recycled aggregate concrete. The properties, characteristics and methods of modification of nanosilica are considered, the mechanical properties and evolution of the microstructure of nanosilica after modification are highlighted. The results show that nanosilica can improve the mechanical properties of recycled aggregate concrete mainly due to the good nucleation effect, filling effect and volcanic ash effect of nanosilica.
Keywords: Nano-silica, Recycled concrete, Mechanical properties

Вплив нанокремнезему на механічні властивості переробленого бетону: огляд. *Yuqing Yan, Fang Qin*

Наведено огляд літератури щодо використання нанокремнезему для покращення поверхні переробленого наносилікатного заповнювача та зміни властивостей бетону з переробленого заповнювача. Розглянуто властивості, характеристики та способи модифікації нанокремнезему, виділено механічні властивості та еволюція мікроструктури нанокремнезему після модифікації. Результати показують, що нанокремнезем може поліпшити механічні властивості бетону з переробленим наповнювачем, що в основному пов’язано з хорошим ефектом утворення зародка, ефектом наповнення та ефектом вулканічного попелу нанокремнезему.

Introduction

With the acceleration of urbanization, the demolition and reconstruction of buildings produce a large amount of construction waste, which brings a huge burden to society and the environment [1]. Recycling/reusing construction and engineering waste is important for sustainable development and has been research focus in recent years. Some of the construction waste processed into recycled aggregate (RA) can effectively replace some of the natural aggregate, which is important for protecting the environment and natural resources; this can not only effectively solve resource problems, but also save construction waste recycling costs and land resources. However, recycled concrete exhibits multiscale properties along with interface complexity. A large number of research results show that with an increase of RA substitution rate, the mechanical properties of recycled aggregate concrete (RAC) gradually decrease [2, 3], Therefore, to ensure the promotion and application of RAC in practical engineering, scientists have carried out research on strengthening RA and RAC [4, 5].

The use of nanomaterials has improved RAC properties at the micron and nanoscale levels, especially by refining the structure of C-S-H and optimizing the microstructure of the matrix and interfacial transition zone (ITZ). Nanomaterials act as bridge connections at the secondary and tertiary levels [6]. With the development of nanotechnology in civil engineering materials, many nanomaterials have been developed and applied in RAC [7, 8]: nanosilica (NS), nano-TiO_2 (NTO), carbon nanotubes (CNT), graphene oxide, and so on. Research on the performance of nanoparticle-modified RACs has made great progress and played various roles in different stages of RAC production. For example, the high specific surface area and nanometer size of NS particles show a unique nucleation effect on the early hydration of cement-based materials [9], but also has a negative impact on workability. In the hydration process, NS particles of volcanic ash will undergo secondary hydration reaction with CH crystals to form a cement-based material microstructure reinforced with C-S-H gel [10]. Therefore, NS has great application prospect in RAC.

Although several review papers have been published to discuss the utilization of NS in cement-based materials [11], no literature has been reported on the application of NS in RAC. In this paper, the effects of different modification methods of NS (direct blending, dipping and spraying) on the mechanical properties and microstructure of RAC are reviewed, including compressive strength, bending strength, tensile strength, bonding strength, elastic modulus and stress-strain, hydration properties and pore structure[12]. This paper will provide a new idea for the practical application of NS in building materials.

1. Properties of nanoparticles

As reported [13], the size of NS is generally ranged 8 nm ~ 40 nm, and it is an amorphous white powder or transparent solution. NS has a nanocrystalline structure composed of SiO_2 molecules, with a short-range sequence of SiO_4 tetrahedrons with shared angles. Due to amorphous structure and random packing of SiO_4, Si ions occupy all the tetrahedral voids because they are smaller than oxygen ions. The SiO_4 tetrahedral is thus the building block of silica particles. Its special three-dimensional chain structure provides a large number of unsaturated bonds and hydroxyl groups for bonding between material groups. NS has attracted great attention in RAC because of its large specific surface area, easy dispersion, good stability and high pozzolanic activity.

2. Mechanical properties

2.1 Compressive strength

Table 1 summarizes the effect of NS on the compressive strength of RAC. It can be found that the RAC strength data is scattered, contradictory and different. The effect of modification by direct doping was studied at NS content in the range of 0.75–8% and RAC growth rate in the range of 2–53%. There are obvious differences in the strength results reported in [13] and [14, 15]. With the same RA content, w/c, curing age and NS content (3wt%), the growth rate of the latter is approximately 3 times higher than that of the former. There are also differences in the optimal NS content from 1 to 6%. It is reported in [14, 16, 17] that with an increase of RA content, the strengthening effect of NS shows a rising trend. Younis et al. [18], however, gave a different result: when the RA content increased from 50% to 100%, the rate of growth of RA strength decreased. Therefore, it is necessary to systematically study and discuss the water-cement ratio or water-binder ratio of RAC, the content and quality of RA, the modification method, type and dosage or concentration of NS, as well as the influence of NS dispersion, size and agglomeration, because these factors have a considerable impact on RAC strength. Although strength results vary widely, the following general rules can be obtained:

(1) The addition of NS is benefit to improve the strength of RAC at different ages, especially at the early age [15, 19-21]. Compared with the control group, the effect in the early stage ranged from 6.7 to 53%, and in the late stage from 1.7 to 28%.[13, 14, 22].

(2) Adding appropriate amount of NS into RAC will increase its compressive strength. If the NS content is too high, RAC tends to decrease in strength and even to have negative growth [23]. This may be due to the fact that under the action of van der Waals force, NS agglomeration becomes more serious [24] and inhibits cement hydration; this partially or completely offsets the enhancement effect of NS [25]. The optimal dosage of NS is mainly 2%-3%.

In addition, some researchers [26, 27] have shown that the spraying method has the best enhancement effect on RAC, followed by impregnation method, and finally additive method. Several scientists also studied the effects of two NS combinations or NS and other SCMs, such as fly ash (FA) and silica fume (SF), on

Table 1 Effects of NS on compressive strength of RAC

Enhance-ment methods	RA content (%)	w/c, w/b#	Dosage (%)	Age (d)	Compressive strength Increment (%)	Optimum content (%)	Refs.
Internal mixing	20	0.28	1, 3, 5	7, 28, 90	About 4-8.5; 5-8.7; 4-8.1	3	[30]
	25	0.4	1.5, 3	7, 28, 90	About 30.5-38; 20.7-29; 17-22.7	3	[14]
	30	0.5	1, 2	7, 28	20.7-24.9; 14.6-21.6	2	
	40	0.28	1, 3, 5	7, 28, 90	About 6-12.4; 8-13.2; 5-11.6	3	[30]
	50	0.4	1.5, 3	7, 28, 90	About 36-44; 23-33.7; 17.4-24.5	3	[14]
	50	0.5	3, 6, 8	28	2.5-8.5	6	[16]
	60	0.28	1, 3, 5	7, 28, 90	About 10-16.4; 6-11.2; 3.5-12	3	[30]
	100	0.3	1, 2, 3	28	3-17	2	[31]
	100	0.4	3	7-365	About 8.5-18	/	[32]
	100	0.4	1, 2	7, 28, 90	2-19; 12-13.2; 2-4.3	2	[22]
	100	0.4	1.5, 3	7, 28, 90	About 43-53; 21.5-37; 18.5-28	3	[14]
	100	0.4	1.5, 3	3, 7, 28	8-45.2; 16.7-37.9; 13.6-27.2	3	[15]
	100	0.4	0.75, 1.5, 3	3, 7, 28	2-15.2; 3-15.7; 4.7-18.5	3	[33]
	100	0.4	0.75, 1.5, 3	7, 28, 90	6.5-12.6; 6.7-17.8; 1.5-9.5	3	[13, 34]
	100	0.4	3	28	17.7	/	[35]
	100	0.5	3,6, 8	28	6.4-12.3	6	[16]
	100	/	1, 2	28	12-21.6	2	[25]
Extra mixing	50	0.48	0.4, 0.8, 1.2	28	10-20.2	1.2	[18]
	50	0.4	1	3-28	4.7-9.2	/	[17]
	70	0.4	1, 2	3, 7, 28	11.6-33.7; 12.3-25.8; 14.4-38.2	2	
	100	0.4	1	3-28	0-14.8	/	[17]
	100	0.4	2	28	20	/	[36]
	100	0.48	0.4, 0.8, 1.2	28	6-16.2	1.2	[18]
	100	0.6 (AS/FA)	1, 2, 3	7	-4-29.5	2	[37]
	100RFA	0.5	1, 2, 3	1, 7, 28	About 4-38; 2-35; 8.9-40	3	[38]
Pre-soak-ing	50	0.5#	2#	7-360	21-38.5	/	[39]
	100	0.56	10.4#	28	About 3.7	/	[26]
	100	0.4	2#	28	25	/	[36]
	100	0.46	2#	28	12.44	/	[40]
	100	0.5	5#	28	14.3	/	[41]
Pre-spray-ing	100	0.56	1-5 (N/A)	28	About 6.0-13.4	3 (N/A)	[26]
	100	0.55	2-8 (N/A)	28	About 0-8.6	4 (N/A)	[27]

the compressive strength of RAC. The results showed that the strength of RAC was further improved by about 14.91-31.7% compared with the single NS modification [22, 28, 29].

Due to the multi-scale nature and complexity of RAC, the mechanism for its strengthening is not fully understood and consensus has not yet been reached. But the action mechanism of NS on RAC is mainly attributed to the filling effect, crystal nucleus effect and volcanic ash effect.

Table 2 Effects of NS on tensile and flexural strength of RAC

Enhancement methods	RA content (%)	w/c, w/b#	Dosage (%)	Age (d)	Split tensile strength and flexural strength Δ, Increment (%)	Optimum content (%)	Refs.
Internal mixing	20	0.28	1, 3, 5	7, 28, 90	About 0-8.4; 2.1-3.3; 1-1.5 (1.3-8.4; 12-14.5; -2.7-7)Δ	3	[30]
	40	0.28	1, 3, 5	7, 28, 90	About -5.3-11; 4.1-5.5; 0-18 (2.6-9.4; 12.6-15.8;0-9.2)Δ	3	
	50	0.5	3, 6, 8	28	-8.2-4.39; (-17.5- -10)Δ	3	[16]
	50	0.5	3, 6, 8	28	-13- -1	3	[42]
	60	0.28	1, 3, 5	7, 28, 90	About -14.7-3; 1.2-2; -3.8-0 (2.7-6.7; 8-15.5; -7-3.5)Δ	3	[30]
	100	0.42	0.5, 1, 2	28	7.89-18.4	1.2	[43]
	100	0.4	0.75, 1.5, 3	28	7.6-16.6; (2.4-14.5)Δ	3	[13, 34]
	100	0.5	3, 6, 8	28	-12.4-4.1; (-8.7- -3.8)Δ	3	[16]
	100	0.5	3, 6, 8	28	-13.45- 2.9	6	[42]
	100	0.6 (AS/FA)	1, 2, 3	7	-3.8-11; (0- 19.4)Δ	2	[37]
Extra mixing	50	0.4	1, 2	7, 28, 56	14.1-34.3; 18.2-15.5; 2.4-17.2	2	[44]
	50	0.48	0.4, 0.8, 1.2	28	4.7-10	0.8	[18]
	70	0.4	1, 2	28	6-20.8	2	[17]
	100	0.48	0.4, 0.8, 1.2	28	1.8-8.16	1.2	[18]
Pre-soaking	50	0.5#	2#	14, 28	42, 44	/	[39]
	100	0.46	2#	28	-4.87	/	[40]

2.2 Flexural strength and tensile strength

According to the results of previous studies, the effects of NS on the tensile strength and flexural strength bending strength of RAC are summarized in Table 2. In general, the addition of an appropriate amount of NS improves the tensile and flexural strength of RAC [13, 34]; the tensile and flexural strength have similar change trends. Regarding the different modification methods, the internal mixing method shows a good effect, but there is less relevant data and it need to be confirmed by further research. The influence range is 1.8-47.54% NS added to enhance RAC. Relatively, the effect of the extra mixing method is quite different. Although most researchers believe [17] that NS doping can promote RAC strength. However, Yonggui et al. [42]and Wang et al. [16] found that the NS doping had negative effects on both the tensile and flexural strength of RAC. After RA was modified by impregnation, the tensile strength of RAC sample at 28d was reduced by 4.87% [40].

2.3 Bond strength

Alhawat et al. [9] studied the effect of NS on the bond strength of steel in RAC under corrosion conditions. The results show that the incorporation of NS improves the bond strength of the steel in RAC. Under non-corrosive conditions, the bond strength of 100RAC-1.5%NS samples is 9%-21% higher than that of the control group. This improvement becomes more significant under corrosive conditions, especially with increasing RA content. After 15 days of corrosion, the bond strength of 100 mm steel in 100RAC-1.5%NS sample increased approximately 3 times. Meng et al. [45] found in a pull-out test on reinforced recycled concrete that after impregnation of RA with a mixed solution of 5%NS+5%NCa and a mixed solution of 5%NCa+1% dispersant, the ultimate stress and bond strength of RAC samples increased by 25.0% and 44.2%, respectively, compared to the control group. This implies increased bond strength between RAC and the steel.

Song et al. [46] investigated the effect of single-doped NS and CNT on the tensile and shear

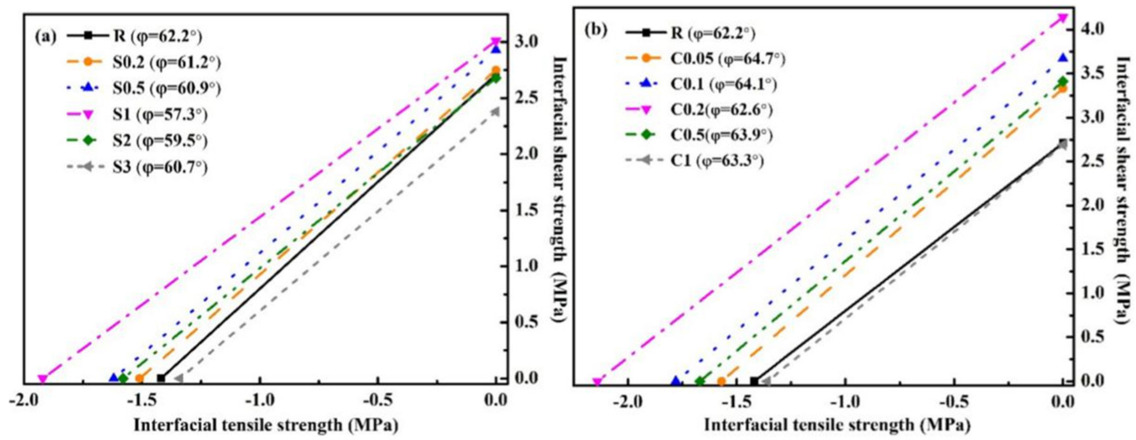

Fig. 1 Interfacial tensile and shear strength interaction diagram of (a) RAC modified with NS; (b) RAC modified with CNTs [46]

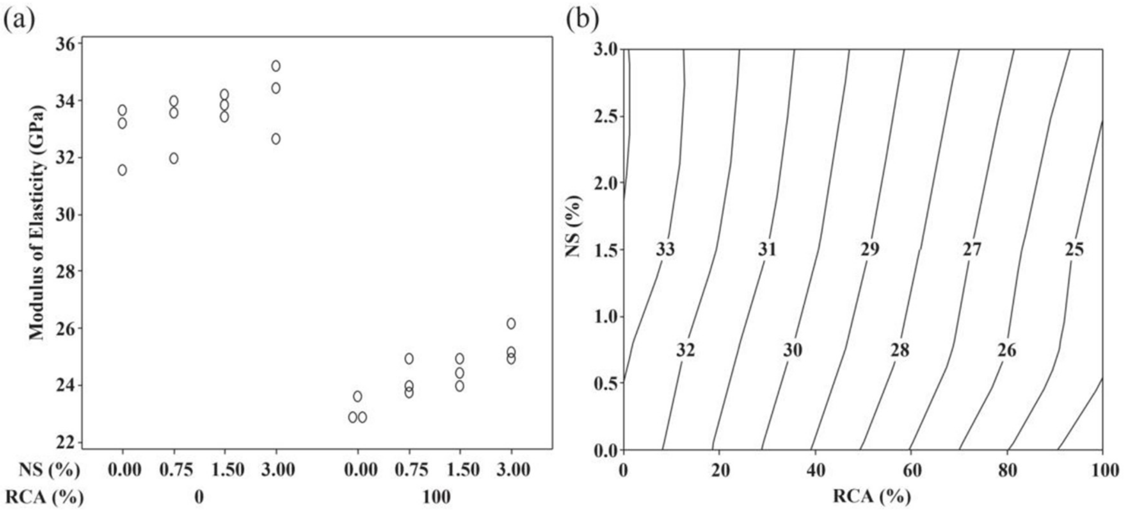

Fig. 2 Modulus of elasticity for RAC containing NS (a) individual plots, (b) contour plots [49]

strength of the RAC interface. The results showed that the interfacial tensile strength of the samples increased by 35% and 51%, respectively, when 1% NS and 0.2% CNT were added to RAC. This is similar to the trend of shear strength. Based on Mohr-Coulomb strength theory [47], he also obtained the interaction diagram of interfacial tensile and shear strength (Fig. 1). It can be seen that the RAC samples containing NS had a slightly smaller internal friction angle (up to 7.9%) than the reference group, and the samples with the addition of CNTs had a slightly larger angle (up to 4.0%). This indicates that the NSs have similar effects on tensile and shear strength.

2.4 Elasticity modulus

Prusty et al. [48] used the Taguchi method to study the effects of cement, RA, NS content and w/c on RAC elastic modulus. The results show that the degree of influence of the above four additives on the elastic modulus of RAC by content is ranked as RA > w/c > cement > NS. Fig. 2 shows that with an increase in the NS content from 0% to 3%, the elastic modulus of the RAC increases from 27.93 GPa to 29.74 GPa [49]. This indicates that NS does not have significant impact on the elastic modulus of RAC, which is consistent with the research results reported in the literature [33]. Li et al. [26] in experiments on modification with RA by spraying and impregnation methods found that the elastic modulus of RAC did not change significantly due to the modification with RA, which is due to the fact that the change in the static elastic modulus mainly depended on the characteristics of aggregate [50].

In contrast to the above, Li et al. [51] found that compared to untreated RAC, the 28d elastic modulus of RAC samples sprayed with NS solution increased by 7.6%; this suggests that

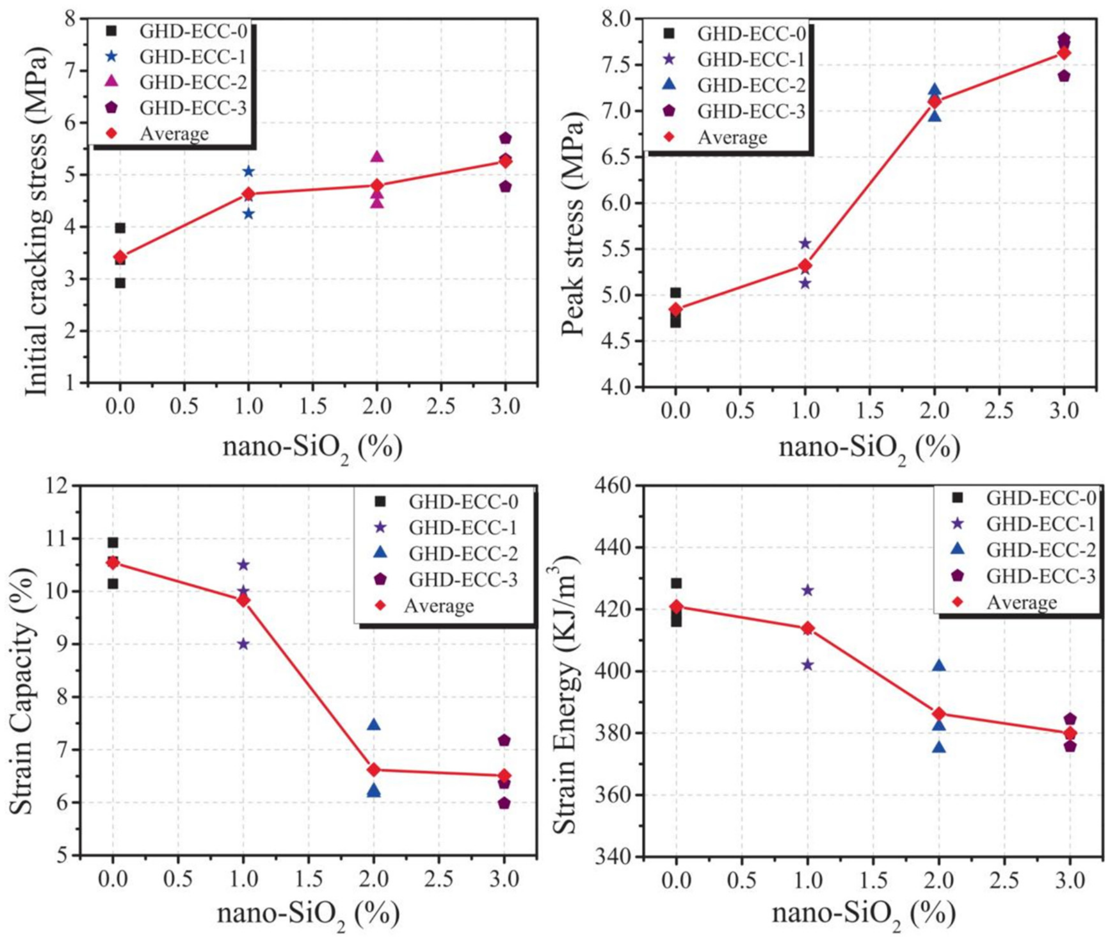

Fig 3. Characteristic parameters of studied GHD-ECCs: (a) initial cracking stress, (b) peak stress, (c) strain capacity, and (d) strain energy [38]

the expansion of the new interfacial transition zone had a better enhancement effect on the elastic modulus of RAC than RA. In addition, Erdem et al. [43] found that adding 0.5%-2%NS into 100%RAC improved the pore structure in the matrix, resulting in a 3%-7% increase in the dynamic elastic modulus of the sample.

2.5 Stress-strain

Moro et al. [52] found in the ductility test that at 500?, mixing 2% NTO into the recycled fine aggregate mortar (RFAM) can improve RFAM ductility values, but the ductility values of samples containing 0.5% and 1% NTO are not satisfactory. Li et al. [25] studied the effect of NS on the dynamic mechanical properties of RAC under impact load. At a low impact velocity of 7.7m/s, RAC-1NS and RAC-2NCA samples show better deformability and ductility. When the impact velocity increases gradually, the decreasing part of the stress-strain curve of RAC is similar. Meanwhile, they also find that NS really reduce the strain rate sensitivity of cement mortar in RAC especially at high dosages of NS and NC. Wang et al. [40] also reported a similar conclusion. Its mechanism is as follows: free water is known to create high adhesive force between crack surfaces of cement mortar at high loading rates, and is better at exhibiting the Stefan effect [53] and inertia effects [54] to delays the onset of excessive cracking [55]. However, the high-water absorption of NS reduces the free water content in RAC.

In contrast to the above, Yunchao et al. [29] based on the stress-strain curve of RAC found that with an increase in the NS content, the average peak strain of the curve almost remained unchanged, indicating that NS had no effect on the stress-strain curve of RAC. Li et al. [51] found that the NS pre-spraying method enhanced ITZ between old mortars, which increased the peak stress of RAC, but had no obvious effect on the peak strain; while the ultimate strain showed a decreasing trend, which meant that the brittleness increased. This is similar to the research results of Wang et al.

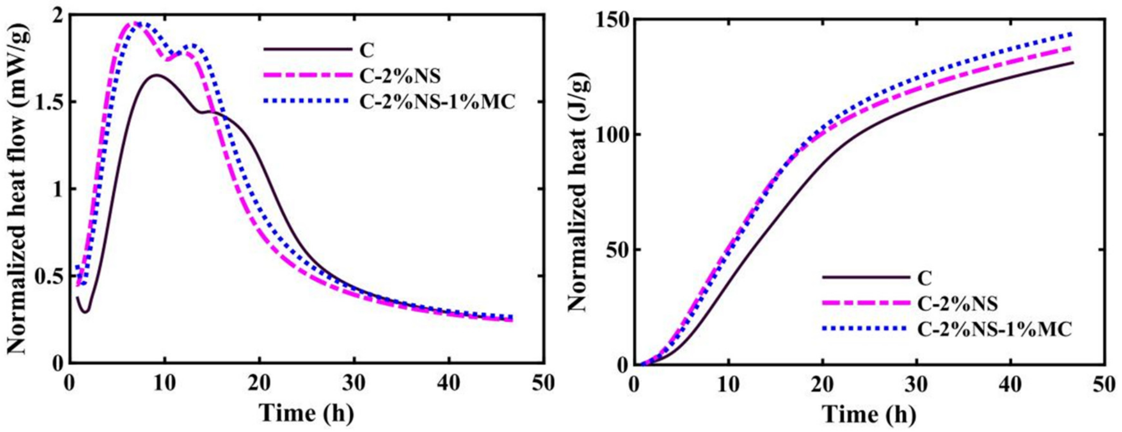

Fig 4. Effects of NS and MC on hydration characteristics of the RAC matrix (a) hydration heat flow; (b) hydration heat [22]

[16] and Mobini et al. [56]. Zhou et al. [38] studied the effect of NS on the performance of green high ductility engineered cementitious composite (GHD-ECC) containing RFA. As shown in Fig 3, the initial cracking stress σ_{tc} and peak stress rise with the increment of NS dosage (Fig. 3a and b). However, the strain capacity ε_{tu} and the strain energy g_{se} display a descending trend with increasing NS content (Fig. 3c and d). For the ECC mixture containing 3% NS, the reduction ratio of ε_{tu} and g_{se} reaches 38.26% and 9.72%, respectively. This indicates that the addition of NS reduces the ductility of ECC with RFA.

In addition to the above mechanical properties, NS also has a positive effect on the dynamic mechanical properties of RAC, such as the storage modulus, loss factor and energy absorption [10, 57, 58].

3. Microstructure

3.1 Hydration characteristics

Previous studies have shown that proper addition of NS can promote cement hydration. As shown in Fig. 4, the incorporation of NS greatly promotes the hydration rate. Compared with a control group, the two peaks of heat flow in the RAC matrix containing NS are significantly earlier, and the peak intensity is greater [22]. Similar phenomena have been observed for other NS [59, 60]. Researchers believe that this phenomenon is mainly due to the following: (1) the nucleation effect of NS promotes the rapid growth of gel on the surface and the "secondary hydration" of the volcano NS and the hydration product CH crystal [61, 62]; (2) the addition of NS promotes the dissolution of aluminum phase of the gel-like material and the deposition of ettringite due to the consumption of calcium sulfate in the pore solution [63]. However, due to the agglomeration problem, too much NS has an inhibitory effect on cement hydration [52, 64].

3.2 Porous structure

The porous structure plays a key role in the performance of RAC. As mentioned above, micropores in cement slurry can be divided into harmless pores (< 20nm), less harmful pores (20-50 nm), harmful pores (50-200 nm) and multi-harmful pores (> 200 nm) according to their sizes [65, 66]; among which pores with a diameter of > 50nm are considered to play a decisive role in the mechanical properties and durability of RAC. The pore diameter < 50nm is closely related to the volume stability of RAC.

Previous studies have shown that the addition of NS reduces the total porosity of the RAC matrix and makes the microstructure denser, thus enhancing the performance of RAC. Mercury injection (MIP) test results confirmed [23] that after adding NS and NL to RAC, the total porosity of the sample was reduced by 24.5% and 26.5%, respectively. It has also been further confirmed in other NS experiments [67]; and the total porosity and specific surface area have similar changing trends [68]. Compared with the additive method, the total porosity of RAC modified by the immersion method is reduced by 45.3% [36]. At the same time, several scientists also obtained similar results using the void volume test. [35, 44].

4. Interfacial transition zone

The study of ITZs changes caused by NS at the nanoscale has aroused great interest among scientists. Due to the complexity and

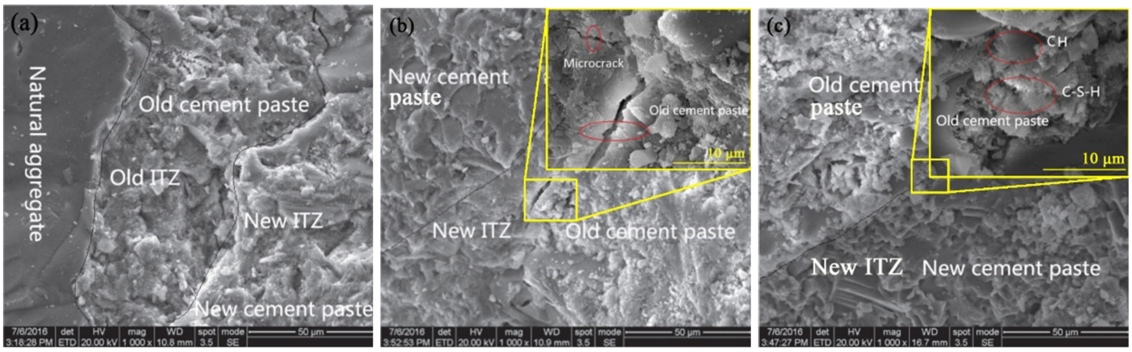

Fig 5. Microstructure of new and old ITZs in RAC modified with NS: (a)RAC0; (b)RAC-NL; (c)RAC-NS[23]

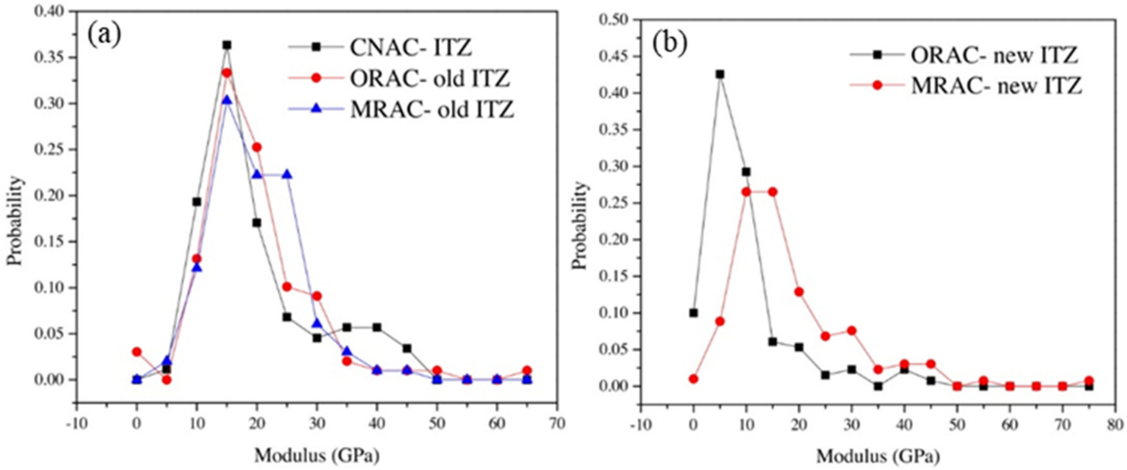

Fig. 6 The elastic modulus of ITZs in the ORAC and MRAC: (a) old ITZs, (b) new ITZs [39]

diversity of ITZs, it is the weakest area of RAC mechanical properties [69], and the gel in this area is prone to cracking, which hinders the load transfer. Therefore, changes in the microstructure and hardness of ITZ will significantly affect the performance of RAC [70, 71]. The SEM analysis results (Fig. 5) shows that the incorporation of NS and nano-Al_2O_3 (NL) makes the ITZ microstructure of RAC denser and reduces the number of microcracks. Compared with NL, NS has a better effect on microstructure improvement. This is related to the poor dispersion of NL, which is difficult to penetrate into ITZ. Mukharjee [32, 35] found that with incorporation of NS, the porosity, unhydrated cement content and micro-hardness of ITZs in RAC were significantly improved, and gradually tended to NAC samples, which was also confirmed by Meng et al. [72].

A lot of reports indicate that NS impregnation can effectively improve the RAC interfacial transition zone [36]. Zhang et al. [39] characterized the changes in the ITZs width and elastic modulus using the nanoindentation method. As can be seen from Fig. 6, there is no significant difference the elastic modulus of ITZ-II between NAC, RAC and modified RAC samples. However, when comparing ITZ-III, the probability distribution of elastic modulus below 7 GPa for ITZ in the ORAC sample is 0.53, while for MRAC it is only 0.1. Moreover, the probability distribution of high elastic modulus of MRAC (10–20 GPa) is much larger than that of ORAC. In conclusion, the elastic modulus of ITZ-III in the RAC sample can be effectively improved by the method of NS immersion, which is consistent with the research results reported in [73]. Zhang et al. [41] and Yue et al. [22] used similar methods to study the modifying effect of NS and the mixed suspension impregnation method of nanocalcium carbonate or micron calcium carbonate on ITZ. The results showed that the two kinds of slurry had little effect on the elastic modulus of ITZ-II, but showed a significant effect on ITZ-III. When testing RA modified by impregnation with a mixed solution of NS and microorganisms, it was found that the ITZ contains thick deposits of calcium carbonate and hydration products, which fill pores and microcracks, thereby strengthening the bonds between the old mortar and new mortar [74].

5. Summary and outlook

This article reviews recent publications on NS-modified RAC in detail, and the main findings can be summarized as follows:

(1) Due to the large specific surface area, high water absorption, poor dispersion and other characteristics of NS, the incorporation of NS into RAC will reduce its workability. However, nano-sized NS particles exhibit good nucleation effect, which promotes the hydration reaction of cement and shortens the setting time.

(2) Many results confirm that the compressive strength, flexural strength, flexural strength and bond strength of RAC are improved by appropriate amount of NS, but the effect on elastic modulus and optimal dosage of NS remains controversial. The enhancement mechanism of NS is mainly attributed to nucleation, filling, volcanic ash effect and bridging effect. Although a small number of studies have shown that the enhancement effect of the impregnation and spray methods is better than that of the admixture method, further research is needed to confirm.

The existing research results confirm that NS-modified RAC has great potential, and its commercial application also has exciting prospects, but further efforts are needed in future research from the following aspects:

(1) The modification effect of NS is limited by dispersion, and agglomeration can have a serious negative impact on RAC performance. Therefore, it is necessary to optimize the technology and scheme for de-agglomeration of NS.

(2) Compared with the mechanical properties, there are relatively few studies on the effects of NS on the durability and volume stability of RAC. Therefore, it is necessary to carry out more comprehensive and in-depth tests on durability and volume stability.

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