

1           **Bond of nanoinclusions reinforced concrete with old concrete:**  
2           **strength, reinforcing mechanisms and prediction model**

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9           **Abstract:**

10   This paper investigated the bond strength of eight nanoinclusions reinforced concrete  
11   with old concrete through a splitting tensile test. The reinforcing mechanisms of bond  
12   due to nanoinclusion was also explored by means of scanning electron microscope and  
13   energy dispersive spectrometer. A prediction model for the bond strength between  
14   nanoinclusion reinforced concrete with old concrete substrate was developed and  
15   calibrated against the experimental results obtained. The experimental results indicated  
16   that bond strength between nanoinclusions reinforced concrete and old concrete can  
17   reach 2.85 MPa, which is 0.8 MPa/39.0% higher than that between new concrete  
18   without nanoinclusions and old concrete. The reinforcing mechanisms can be attributed  
19   to the enrichment of nanoinclusions in the new-to-old concrete interface, compacting  
20   the interfacial microstructures and connecting hydration products in micropores of old  
21   concrete with that in bulk new concrete. In addition, the prediction model proposed on  
22   the basis of reinforcing mechanisms can accurately describe the relationship of the  
23   nanoinclusion content and the bond strength of nanoinclusions reinforced concrete with  
24   old concrete.

25   **Keywords:** Nanoinclusions reinforced concrete; New-to-old concrete; Bond strength;  
26   Reinforcing mechanisms; Prediction model  
27  
28

## 29 1 Introduction

30 A large number of concrete structures cannot reach their design service life due to  
31 damage caused by the combined effects of mechanical loads and environmental factors,  
32 for example fatigue, shrinkage, creep, temperature change, freeze-thaw weathering, etc.  
33 In order to prolong the service life of deteriorated concrete structures, repair and  
34 rehabilitation using new concrete is generally adopted. However, it is estimated that  
35 about half of concrete repairs fail; most of which can be attributed to debonding at the  
36 new-to-old concrete interfaces, confirming the importance of reliable bonding between  
37 new and old concretes [1-3].

38 Bond strength is a key indicator, representing the quality of interface between new  
39 and old concretes. Such new-to-old concrete bond strength is mainly governed by  
40 mechanical interlock and van der Waals forces, which is influenced by interface  
41 roughness (affecting mechanical interlock), the shrinkage difference of new and old  
42 concrete (affecting interfacial internal stress state), the microstructures of the new-to-  
43 old concrete interface (affecting van der Waals forces), etc. [4, 5]. Based on the factors  
44 governing the bond strength between new and old concretes, previous research  
45 investigations have taken two directions to improve such bond strength. The first one  
46 is focused on preparation techniques to increase the surface roughness of old concrete,  
47 therefore, dramatically enhancing the mechanical interlock between new and old  
48 concretes. It is reported that the bond strength between new and old concretes with  
49 rough surface is several to over ten times higher than that with smooth surface [6-8].  
50 However, this method is difficult to be applied in repairs of inherent structural cracks.  
51 The second technique is mainly concerned with the development of improved repair  
52 materials that are volumetrically stable, i.e. undergo neither shrinkage nor expansion  
53 once installed, and would display compatible modulus of elasticity, strength, creep,

54 shrinkage, thermal expansion, permeability and electrochemical characteristics to the  
55 substrate existing concrete. For example, fibers, mineral additives, and shrinkage  
56 reducing agents are incorporated so as to limit the shrinkage difference between new  
57 and old concrete [9-12]. Meanwhile, nanoinclusions have been certificated to be  
58 effective in improving the mechanical properties and durability of concrete, through  
59 densifying microstructures and reducing shrinkage of concrete [13-19]. This is  
60 beneficial to achieve reliable bonding between nanoinclusions reinforced concrete and  
61 old concrete, showing a promising material for concrete rehabilitation and  
62 strengthening [20-26]. The observed best modification of bond strength between  
63 nanoinclusions reinforced concrete and old concrete with smooth surface includes a  
64 29.5% increase in tensile strength [21] and a 21.6% increase in slant shear strength [22].  
65 However, there is a lack of broader research with regard to the quantification of bond  
66 strength between different types of nanoinclusions reinforced concrete and old concrete,  
67 a better understanding of reinforcing mechanisms to further control the repair effects of  
68 nanoinclusions reinforced concrete and development of prediction models of bond  
69 strength between nanoinclusions reinforced and old concretes.

70 Therefore, this paper aims to investigate the bond strength, reinforcing mechanisms  
71 and prediction model of nanoinclusions reinforced concrete with old concrete. A  
72 splitting tensile test was carried out to determine the bond strength between **eight**  
73 different types of nanoinclusions reinforced concrete and old concrete as well as the  
74 splitting tensile strength of concrete with nanoinclusions. After the splitting tensile test,  
75 scanning electron microscope (SEM) and energy dispersive spectrometer (EDS) were  
76 performed in order to explore the reinforcing mechanisms. Finally, a prediction model  
77 of bond strength between nanoinclusions reinforced concrete and old concrete was  
78 established.

## 79 2 Experimental programs

### 80 2.1 Materials and mix design

81 Due to their small size, nanoinclusions can only affect concrete in a small range  
82 around them, but cannot fill the larger pores [27, 28]. Concrete with compact  
83 microstructure may fully achieve the reinforcing effect of nanoinclusions. Therefore,  
84 reactive powder concrete was selected in this study where the modified Andreasen and  
85 Andersen packing model [29] is used for mix design of concrete, as presented by  
86 Formula 1 below.

$$P(D) = \frac{D^q - D_{min}^q}{D_{max}^q - D_{min}^q} \quad (1)$$

87 where  $D$  is the particle size of solid particles in concrete;  $D_{max}$  and  $D_{min}$  are the  
88 maximum and minimum particle size;  $q$  is the distribution modulus ranging from 0 to  
89 0.28 according to the previous researches [30-32], and its value in this study is equal to  
90 0.28;  $P(D)$  is the fraction of the total solid particles being smaller than  $D$ .

91 Ordinary Portland cement with a strength of 42.5 R is employed for all specimens  
92 prepared in this study. Quartz sand with a size range of 0.12-0.83 mm is used as  
93 aggregate. The polycarboxylate superplasticizer has a reducing water capability to an  
94 extent of 30%. In addition, class II fly ash and silica fume with the average particle size  
95 of 0.15  $\mu\text{m}$  are used as mineral mixtures in this study. Eight different types of  
96 nanoinclusions are selected to reinforce the new concrete according to previous studies  
97 [33]; the properties of which are listed in Table 1. The contents of different  
98 nanoinclusions are determined on the basis of the previous literature [33]. The mix  
99 proportions of concrete with different types of nanoinclusions are listed in Table 2.

100 Table 1. Properties of nanoinclusions

Types	Abbreviation	Purity (%)	Diameter (nm)	Length ( $\mu\text{m}$ )	Thickness (nm)	Density ( $\text{g}/\text{cm}^3$ )	Specific surface area
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							(m <sup>2</sup> /g)
Nano silica	S	≥99	20	-	-	2.2	≥600
Silica-coated rutile titania	T	≥96	20	-	-	4	-
Monoclinic zircon	Z	≥99	20	-	-	5.8	≥25
CNTs	CNTs	-	20-30	0.5-2	-	2.1	>120
Hydroxyl functionalized CNTs	H-CNTs	-	<8	0.5-2	-	2.1	>380
Nickel coated CNTs	Ni@CNTs	-	20-30	10-30	-	6.2	70
Multi-layer graphene	MLG	-	<2000	-	1-5	2.25	500
Nano BN	NB	99.9	120	-	5-100	2.3	19

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102

Table 2 The mix proportions of nanoinclusions reinforced concrete

Nanoinclusion	Code	Mix proportions (mass ratio)						Superplasticizer (%)
		Cement	Nanofillers	Fly ash	Silica fume	Aggregate	Water	
Control mix	Blank*	1	-	0.25	0.313	1.375	0.375	1.5
S	S-1	0.99	0.01					2.0
	S-2	0.98	0.02	0.25	0.313	1.375	0.375	2.5
	S-3	0.97	0.03					3.0
T	T-1	0.99	0.01					
	T-2	0.98	0.02	0.25	0.313	1.375	0.375	1.5
	T-3	0.97	0.03					
Z	Z-1	0.99	0.01					
	Z-2	0.98	0.02	0.25	0.313	1.375	0.375	1.5
	Z-3	0.97	0.03					
CNTs	CNTs-0.1	0.999	0.001					
	CNTs-0.3	0.997	0.003	0.25	0.313	1.375	0.375	1.5
	CNTs-0.5	0.995	0.005					
H-CNTs	H-CNTs-0.1	0.999	0.001					
	H-CNTs-0.3	0.997	0.003	0.25	0.313	1.375	0.375	1.5
	H-CNTs-0.5	0.995	0.005					
Ni@CNTs	Ni@CNTs-0.1	0.999	0.001					
	Ni@CNTs-0.3	0.997	0.003	0.25	0.313	1.375	0.375	1.5
	Ni@CNTs-0.5	0.995	0.005					
MLG	MLG-0.1	0.999	0.001					
	MLG-0.3	0.997	0.003	0.25	0.313	1.375	0.375	1.5
	MLG-0.5	0.995	0.005					
NB	NB-0.1	0.999	0.001					
	NB-0.3	0.997	0.003	0.25	0.313	1.375	0.375	1.5
	NB-0.5	0.995	0.005					

103 \* Blank is used for the old concrete and the new concrete without nanoinclusions.

104

## 105 2.2 Specimen preparation

106 In this study, specimens with scale-up interface were fabricated, the geometric  
107 dimension of which is exhibited in Figure 1 (a). The concrete blocks with size of 40  
108 mm×40 mm×80 mm were, initially, cured in water for 28d and, then, in air for 365d.

109 These concrete blocks were used as old concrete to fabricate the specimens with new-  
 110 to-old concrete interface. The key issue in specimen preparation is to homogeneously  
 111 disperse nanofillers in RPC [34, 35]. Poor dispersion of nanofillers weakens the  
 112 nanofiller modification effect or even acts as defects in concrete. This study used  
 113 polycarboxylate superplasticizer as dispersing agent. Additionally, stirring and  
 114 ultrasonic were applied for dispersing non-carbon nanofillers (namely nano silica, nano  
 115 titania, nano zircon, and nano BN) and carbon nanofillers (namely CNTs, H-CNTs,  
 116 Ni@CNTs, and multi-layer graphene). Using these dispersing methods, different  
 117 nanofillers can be evenly distributed in concrete matrix [36, 37]. The detailed  
 118 fabrication process of specimens refers to previous examples from the literature [33].  
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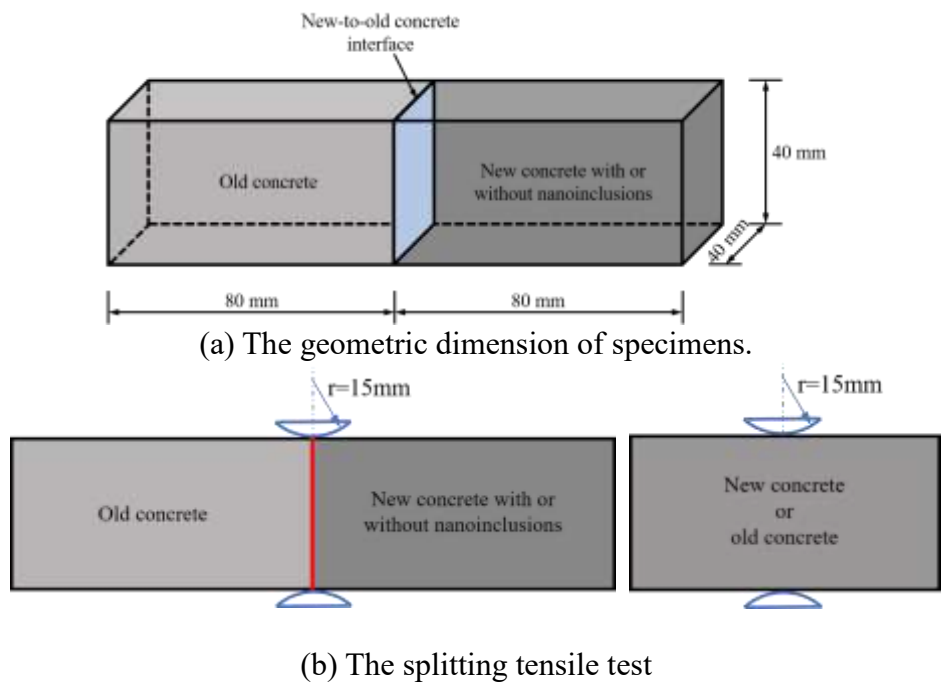


Figure 1. Experimental diagrams

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### 121 2.3 Measurements

122 According to Chinese National Standard GB/T 50081-2019 [38], the splitting tensile test  
 123 was performed on six specimens in each group to evaluate the bond strength of  
 124 nanoinclusions reinforced concrete with old concrete, as shown in Figure 1 (b). The

125 average value of the bond strengths of 6 specimens in each group was recorded as the  
126 final bond strength. The test specimens were loaded to failure at a displacement control  
127 rate of 0.02 mm/min. The splitting tensile strength  $f_t$  is calculated by Formula 2 [38].

$$f_t = \frac{2P}{\pi A} \quad (2)$$

128 where  $P$  is the maximum applied load, and  $A$  ( $=1600 \text{ mm}^2$ ) is the area of the bonding  
129 plane. In addition, the splitting tensile test was also performed on old concrete or  
130 nanoinclusions reinforced concrete to characterize the splitting tensile strength for each  
131 material alone, as shown in Figure 1 (b).

132 After the splitting tensile test, SEM and EDS were performed to observe the  
133 microstructure and morphology of the hydration products on the two failure surfaces  
134 (old concrete side and nanoinclusions reinforced concrete side) of each group.  
135 Moreover, the EDS mapping analysis was carried out on titanium, zirconium, and  
136 nitrogen that are different from the components of cement and the elements introduced  
137 in sample pretreatment.

### 138 3 Results and discussions

#### 139 3.1 Bond strength

140 In this experiment, all specimens were failed along the interface between  
141 nanoinclusions reinforced concrete and old concrete, as shown in Figure 2. Therefore,  
142 the bond strength can be calculated by Formula 2. Figure 3 demonstrates the bond  
143 strength between new concrete with/without nanoinclusions and old concrete,  
144 indicating that the bond strength of nanoinclusions reinforced concrete with old  
145 concrete is higher than that of new concrete without nanoinclusions with old concrete.  
146 For nanoparticles, the bond strengths of specimens with 1 wt.% of S, 3 wt.% of T, and  
147 2 wt.% of Z are 2.38 MPa, 2.62 MPa, and 2.43 MPa, respectively; while that of the

148 specimen without nano inclusions is 2.05 MPa. As for nanotubes, the incorporation of  
 149 0.3 wt.% of CNTs, 0.3 wt.% of H-CNTs, and 0.5 wt.% of Ni@CNTs in the new concrete  
 150 makes the bond strength reach 2.54 MPa, 2.57 MPa, and 2.76 MPa, respectively.  
 151 Similarly, when 0.5 wt.% of MLG and 0.5 wt.% of NB are added to the new concrete,  
 152 the bond strengths reach the maximum value of 2.85 MPa and 2.76 MPa. By  
 153 comparison, bond strengths of nano inclusions reinforced concrete and old concrete in  
 154 this study are superior to previous research [21-24].

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Figure 2. Representative failure form

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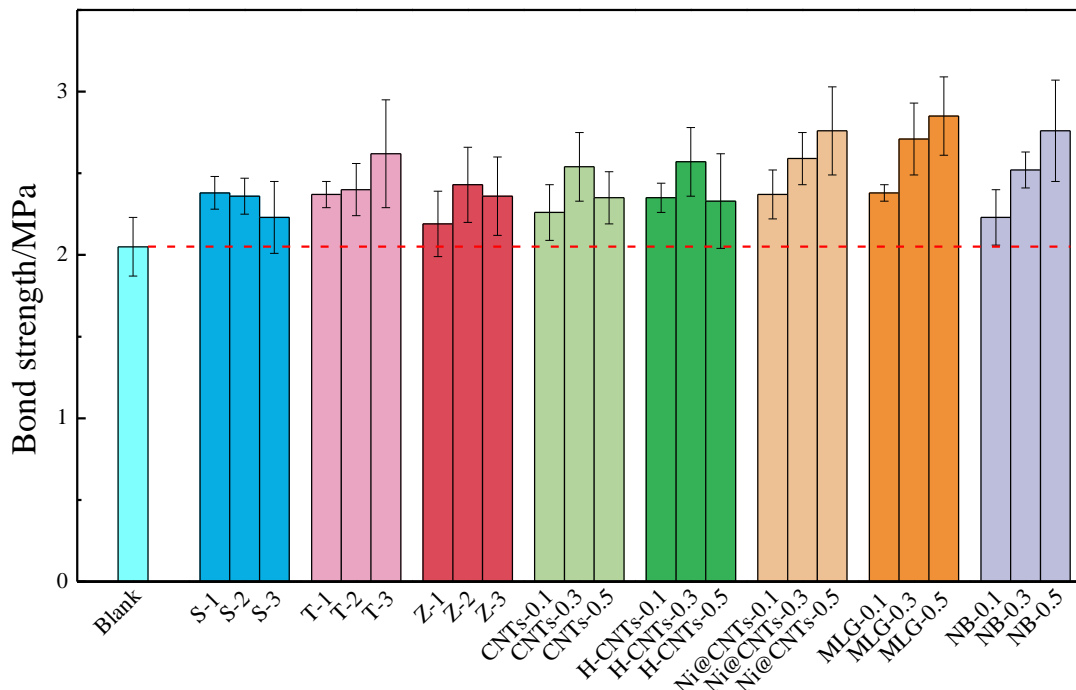


Figure 3. The bond strength between new concrete with/without nano inclusions and old concrete



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160 Figure 4 illustrates the splitting tensile strength of new concrete with/without  
161 nanoinclusions and old concrete. The experimental results show that the splitting tensile  
162 strength of concrete can be enhanced by adding nanoinclusions. In addition, the  
163 splitting tensile strength increases with the contents of nanoinclusions. Comparing with  
164 concrete without nanoinclusions, the splitting tensile strengths of concrete achieve the  
165 highest relative/absolute increases of 1.59 MPa/45.2%, 1.32 MPa/37.5%, and 1.26  
166 MPa/35.8% when 3 wt.% of S, T, and Z are added respectively. As for nanotubes,  
167 concrete with 0.5 wt.% H-CNTs shows the highest splitting strength, achieving an  
168 increase of 1.84 MPa/52.3%. Meanwhile, the presence of 0.5 wt.% of CNTs and  
169 Ni@CNTs can maximally increase the splitting strength of concrete by 1.65 MPa/46.9%  
170 and 1.41 MPa/40.1%, respectively. In addition, concrete with 3 wt.% of MLG and NB  
171 shows the greatest increase of 1.60 MPa/45.5% and 1.20 MPa/34.1% in splitting  
172 strength. The splitting strength of old concrete is 0.57 MPa/16.2% higher than the new  
173 concrete without nanoinclusions due to strength development after 28d.

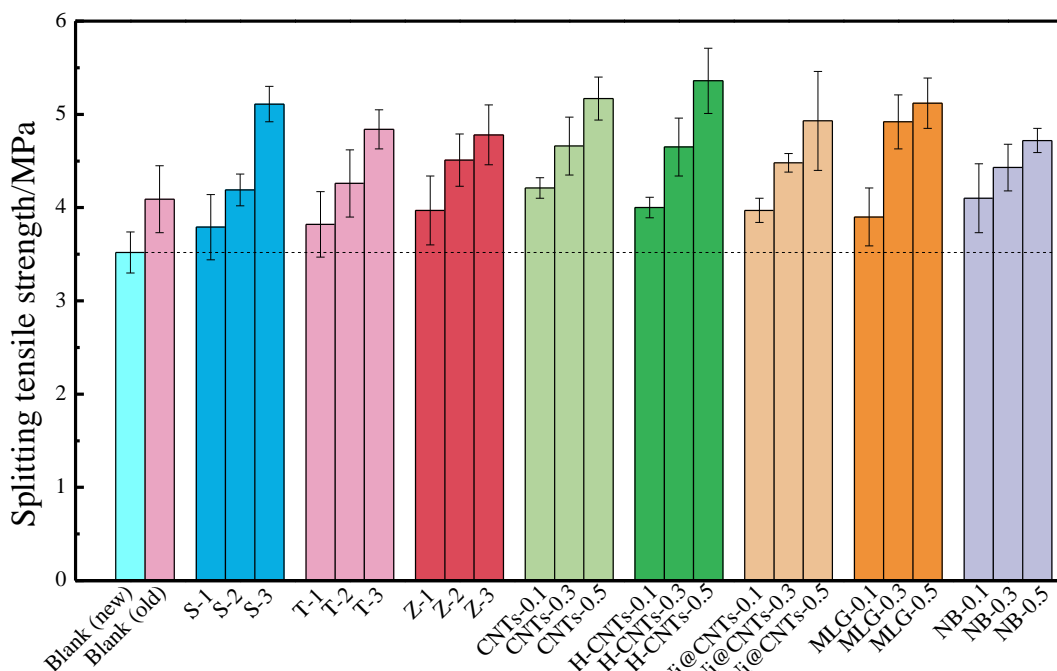
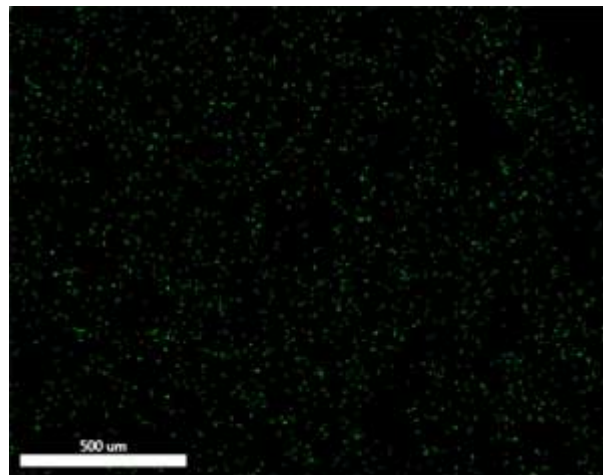


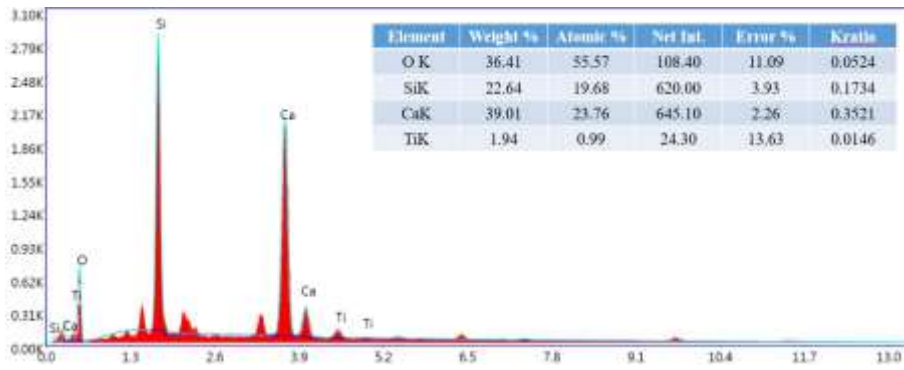
Figure 4. The splitting tensile strength of new concrete without/with nanoinclusions and old

## 174 3.2 Reinforcing mechanisms for bond strength

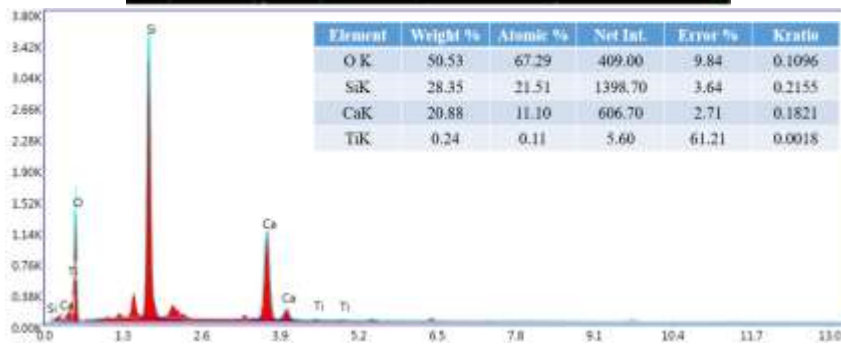
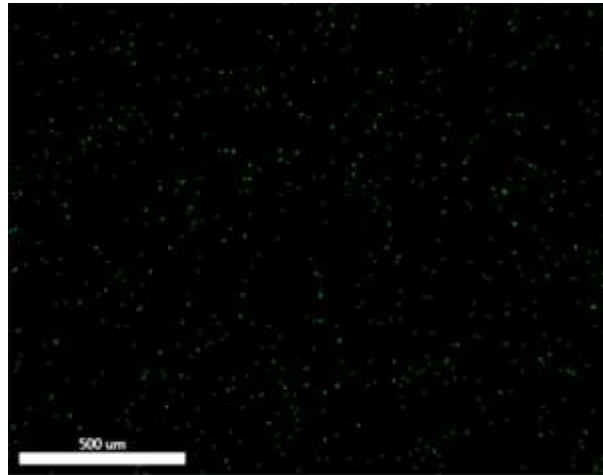
175 The reinforcing mechanisms for bond strength of nanoinclusions reinforced concrete  
176 with old concrete are revealed by SEM observations and EDS analyses. In this study,  
177 EDS mapping analysis is performed on the interior and interface of the nanoinclusions  
178 reinforced concrete in order to characterize the distribution of nanoinclusions in  
179 concrete. For this purpose, the images are firstly converted into gray-scale images, and  
180 the average gray value is calculated [39]. It can be deduced that the average gray value  
181 of the EDS mapping image is larger when there are more nanoinclusions in the analysis  
182 area. The calculated average gray value of concrete interior with 3 wt.% of T is 1.02,  
183 while that of concrete interface is 2.12, indicating that the quantity of T in new-to-old  
184 concrete interface is about 2.08 times larger than that in the nanoinclusions reinforced  
185 concrete interior. Similarly, the quantity of Z and NB in the new-to-old concrete  
186 interface are 1.47 and 1.37 times larger than that in the concrete interior, respectively.

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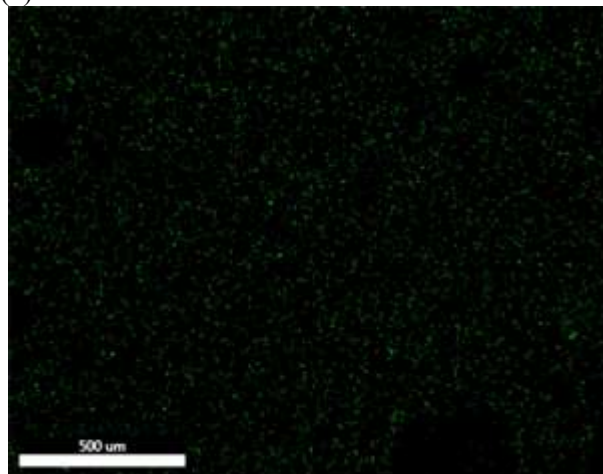


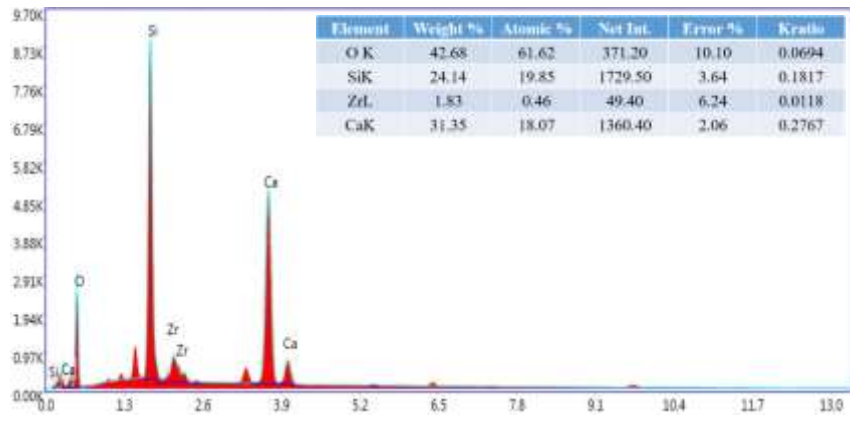


(a) Distribution of titanium in the new-to-old concrete interface

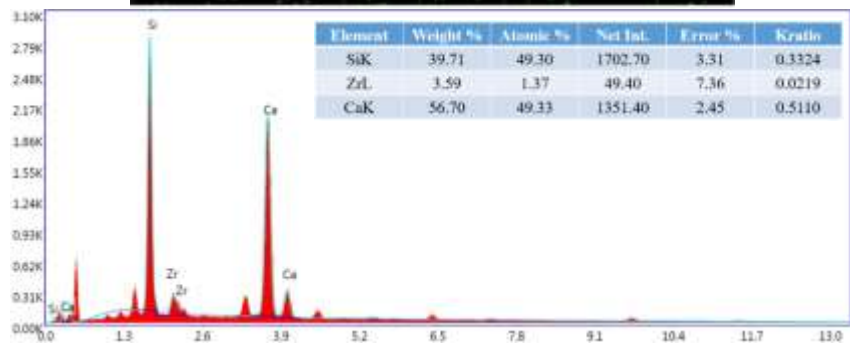
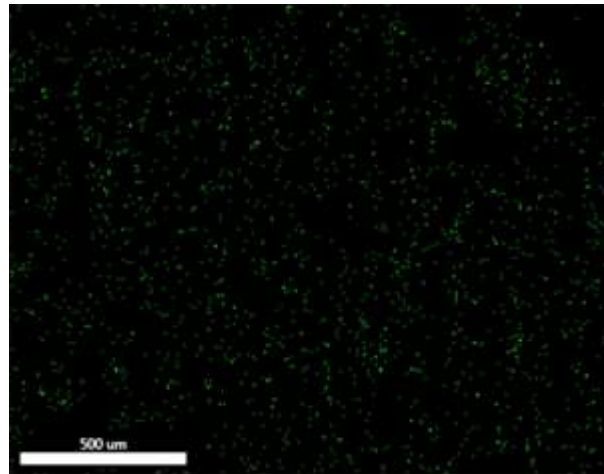


(b) Distribution of titanium in the new concrete interior

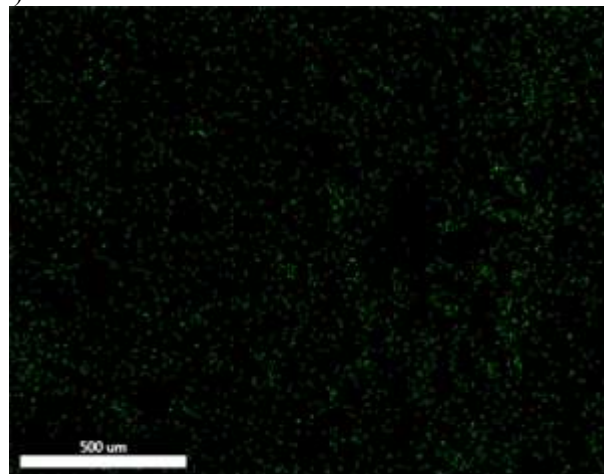


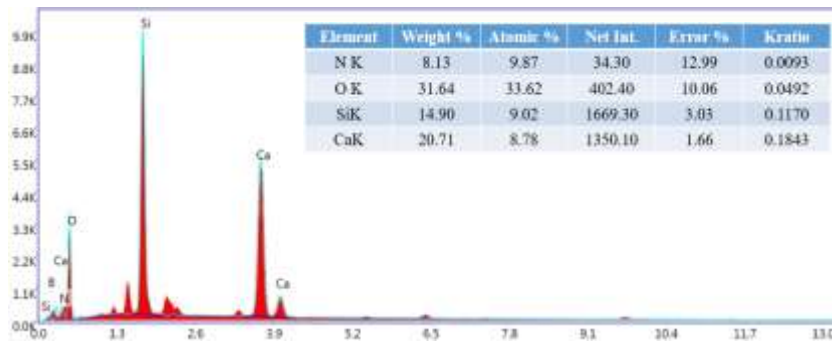


(c) Distribution of zirconium in the new-to-old concrete interface

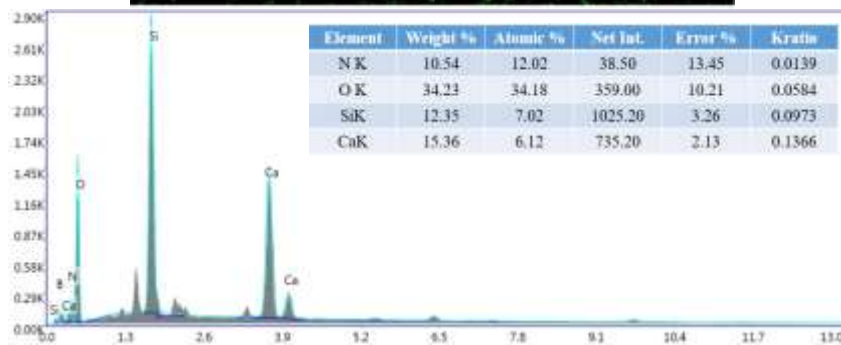
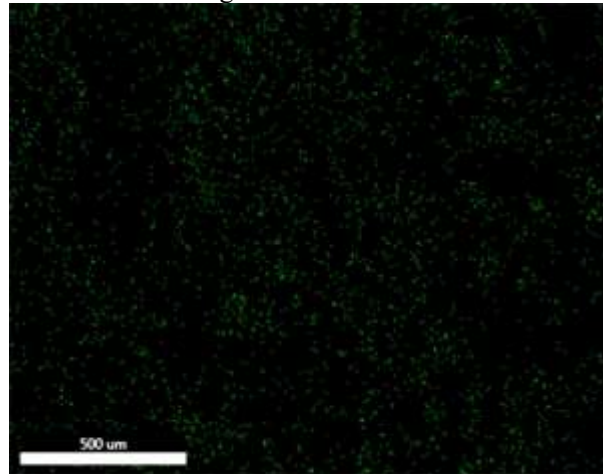


(d) Distribution of zirconium in the new concrete interior





(e) Distribution of nitrogen in the new-to-old concrete interface



(f) Distribution of nitrogen in the new concrete interior

Figure 4. The results of EDS mapping analyses on the new concrete with 3 wt.% of T, 2 wt.% of Z, and 0.5 wt.% of NB as well as the new-to-old concrete interface

188 The EDS mapping results show that nanoinclusions enrich in the new-to-old concrete  
 189 interface as explained by the wall effect and nanoinclusion migration effect, as shown  
 190 in Figure 6. The smaller particles in fresh concrete firstly transfer toward old concrete  
 191 surface while the larger particles move away from old concrete surface due to the wall  
 192 effect, resulting in the phenomenon called scale separation. Afterwards, water is  
 193 absorbed by old concrete and migrates toward the old concrete surface, causing  
 194 nanoinclusions migrating with water in the voids among non-nano particles, and finally  
 195 forming a nanoinclusion enrichment layer in the new-to-old concrete interface.

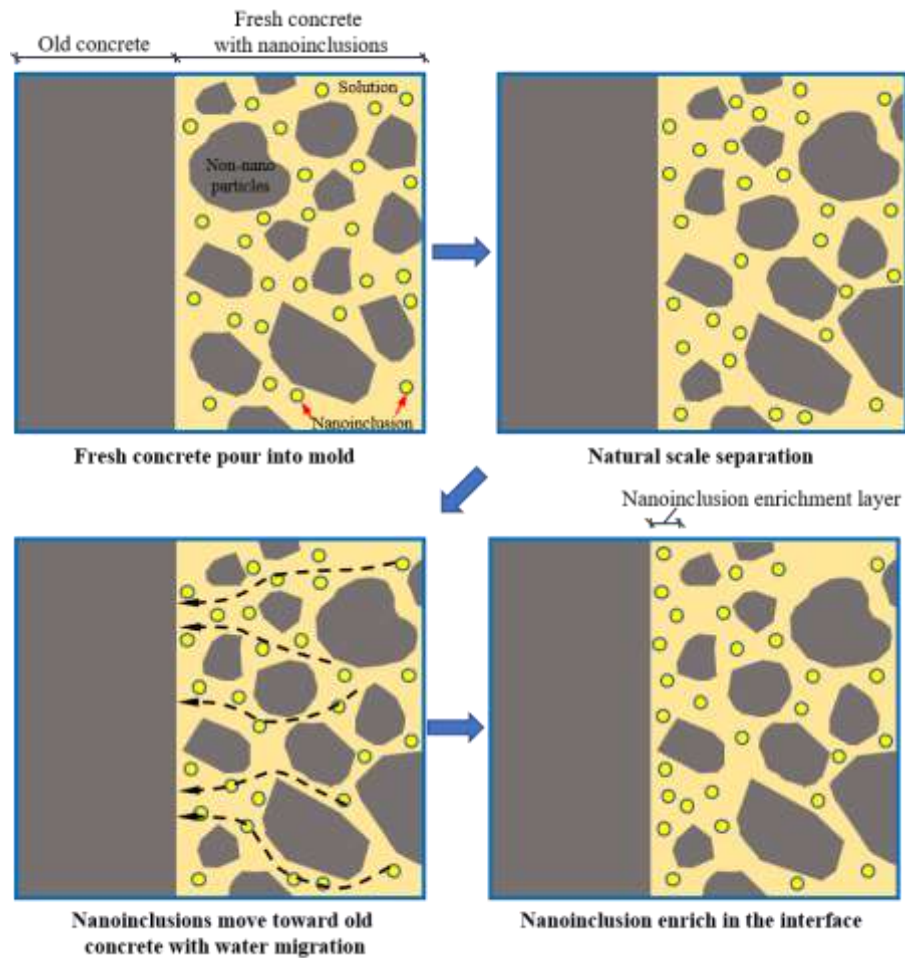


Figure 6. The formation process of nano-inclusion enrichment layer in the new-to-old concrete interface (non-nano particles include aggregates, cement particles, fly ash particles and silica fume particles)

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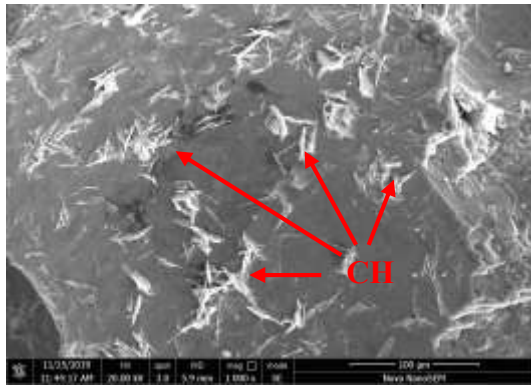
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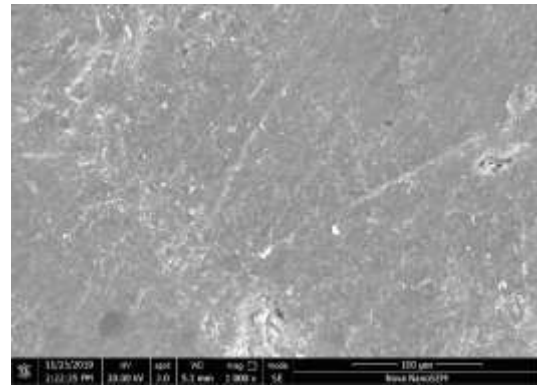
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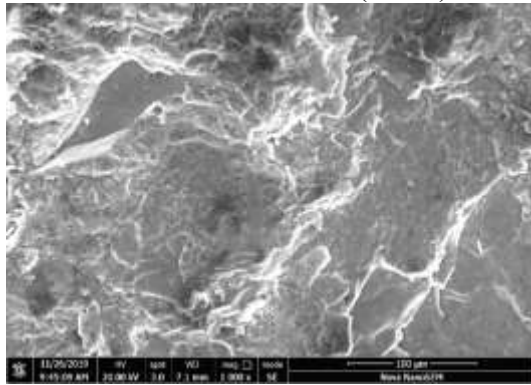
Figure 7 exhibits the original microstructures of the hydration products on the fracture surfaces of nano-inclusions reinforced concrete. It is observed that a large amount of oriented calcium hydroxide (CH) crystal appears on the fracture surface of new concrete without nano-inclusions. In contrast, the microstructures of new concrete with 3 wt.% of T, 1 wt.% of S, 0.5 wt.% of Ni@CNTs, 0.5 wt.% of MLG, and 0.5 wt.% of NB are more compact, showing no obvious CH crystal.



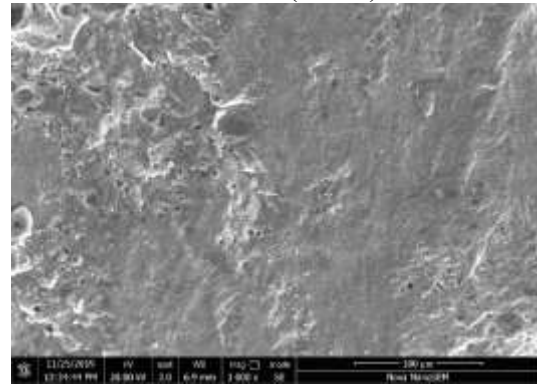
(a) Microstructures of the hydration products on the fracture surface of new concrete without nanoinclusions (1000×)



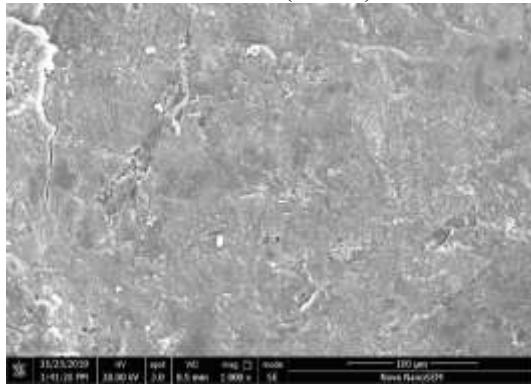
(b) Microstructures of the hydration products on the fracture surface of new concrete with 3 wt.% of T (1000×)



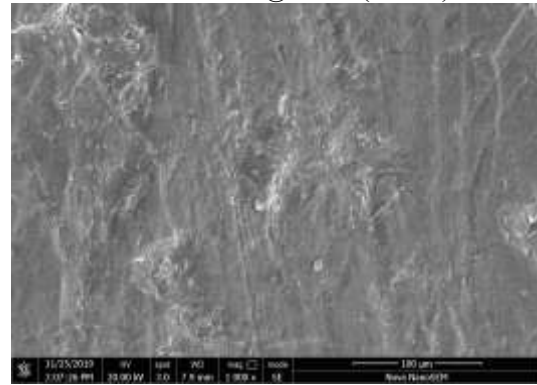
(c) Microstructures of the hydration products on the fracture surface of new concrete with 1 wt.% of S (1000×)



(d) Microstructures of the hydration products on the fracture surface of new concrete with 0.5 wt.% of Ni@CNTs (1000×)



(e) Microstructures of the hydration products on the fracture surface of new concrete with 0.5 wt.% of MLG (1000×)



(f) Microstructures of the hydration products on the fracture surface of new concrete with 0.5 wt.% of NB (1000×)

Figure 7. Microstructures of the hydration products on the fracture surface of new concrete without/with nanoinclusions

205 Figure 8 shows the morphology of the calcium silicate hydrates (C-S-H) gels on the  
 206 fracture surfaces of old concrete. A lot of micropores are found on the old concrete  
 207 fracture surface of the specimens without nanoinclusions. Conversely, the addition of  
 208 nanoinclusions allows the hydration products of nanoinclusions reinforced concrete to

209 fill into the micropores of the old concrete, therefore, no obvious micropores are  
210 observed on the fracture surfaces of old concrete.

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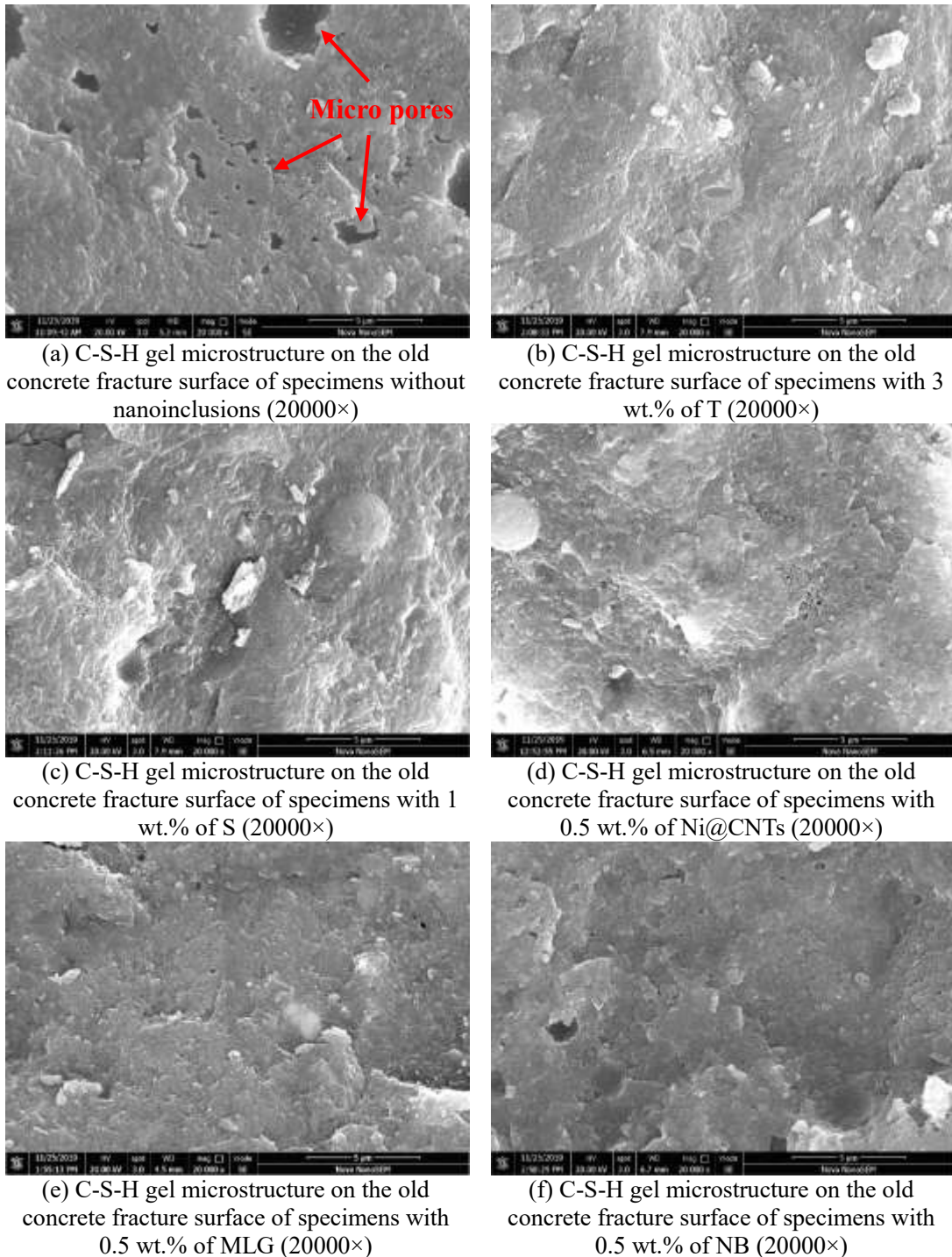
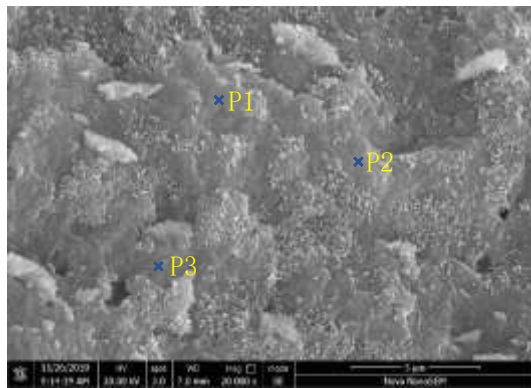


Figure 8. The microstructures of C-S-H gel on old concrete surface

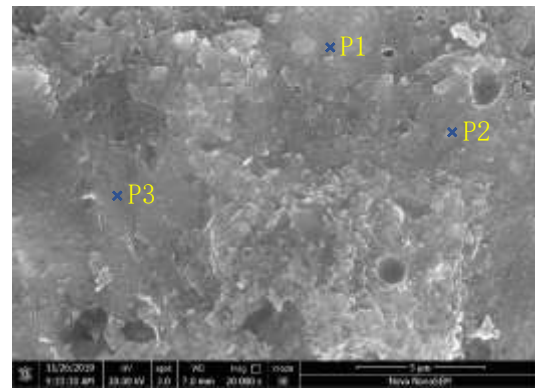
212 Figure 9 illustrates the morphology of C-S-H gels on the fracture surface of new  
213 concrete with nanoinclusions. The C-S-H gels on the fracture surface of concrete



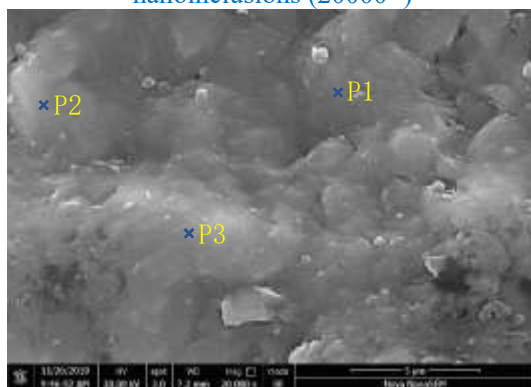
214 without nanoinclusions are loose and porous, containing a lot of CH crystals. Besides,  
215 the average molar ratio of CaO to SiO<sub>2</sub> (Ca/Si ratio) of the C-S-H gel on the surface of  
216 concrete without nanoinclusions is 0.70, as listed in Table 3. On the contrary, the  
217 presence of nanoinclusions makes the C-S-H gels more uniform and improves the  
218 compactness, meanwhile, the Ca/Si ratio of C-S-H gels increases to 0.84-1.02 except  
219 for the incorporation of S. Meanwhile, the Ca/Si ratio of C-S-H gels on the fracture  
220 surface of new concrete with 1 wt.% of S reduces to 0.59, which may be caused by the  
221 enrichment of S in the new-to-old concrete interface.  
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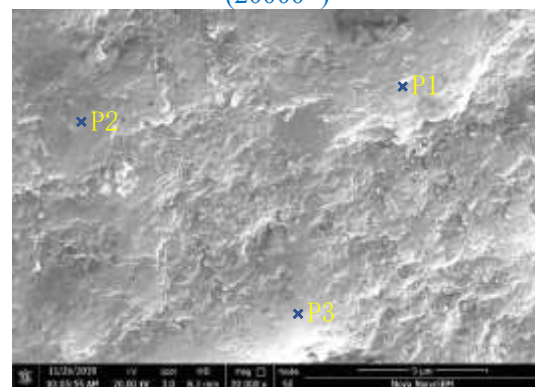
(a) C-S-H gel microstructure on the fracture surface of new concrete without nanoinclusions (20000×)



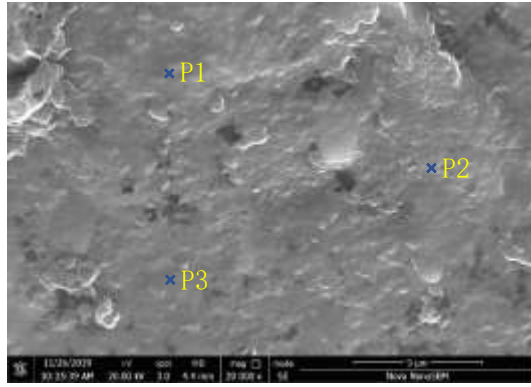
(b) C-S-H gel microstructure on the fracture surface of new concrete with 3 wt.% of T (20000×)



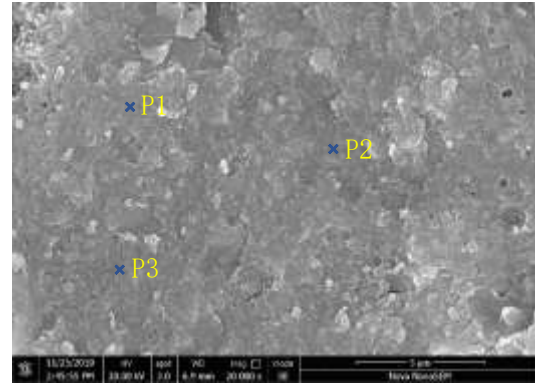
(c) C-S-H gel microstructure on the fracture surface of new concrete with 1 wt.% of S (20000×)



(d) C-S-H gel microstructure on the fracture surface of new concrete with 0.5 wt.% of Ni@CNTs (20000×)



(e) C-S-H gel microstructure on the fracture surface of new concrete with 0.5 wt.% of MLG (20000×)



(f) C-S-H gel microstructure on the fracture surface of new concrete with 0.5 wt.% of NB (20000×)

Figure 9. C-S-H gel microstructure on the fracture surface of new concrete without/with nano inclusions

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Table 3. Ca/Si ratio of C-S-H gel on the fracture surface of new concrete with nano inclusions

Code	EDS analysis point*	Ca (Atomic %)	Si (Atomic %)	Ca/Si ratio	Average Ca/Si ratio
Blank	P1	13.98	18.13	0.77	0.72
	P2	14.89	20.80	0.71	
	P3	14.31	21.24	0.67	
T-3	P1	13.94	20.09	0.69	0.84
	P2	14.88	15.73	0.95	
	P3	14.22	16.37	0.87	
S-1	P1	12.98	21.05	0.62	0.59
	P2	14.88	25.62	0.58	
	P3	9.83	17.59	0.56	
Ni@CNTs-0.5	P1	18.02	16.47	1.09	1.02
	P2	12.51	13.98	0.89	
	P3	20.72	19.52	1.06	
MLG-0.5	P1	21.53	21.37	1.01	1.02
	P2	25.08	22.89	1.10	
	P3	18.19	18.86	0.96	
NB-0.5	P1	15.26	14.44	1.06	0.98
	P2	11.86	12.12	0.98	
	P3	14.7	16.08	0.91	

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\* The EDS analysis point refers to the markings in Figure 9.

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The bond strength of new-to-old concrete interface depends on the combination of chemical bonding (namely ionic bonding and covalent bonding), physical bonding (related to the van der Waals and surface tension forces), and mechanical interlock (concerned with interpenetration of concrete into roughness and porosity of old concrete) [1, 40, 41]. However, the chemical bonding in new-to-old concrete is so weak that can be ignored. As aforementioned, a nano inclusion enrichment layer forms in the new-to-old concrete interface, which can notably improve the physical bonding and

234 mechanical interlock between new and old concrete.

235 The nano-inclusions can notably improve the physical bonding between  
236 nano-inclusions reinforced concrete and old concrete. Previous studies reported that  
237 nano-inclusions in concrete can form numerous nano-core-shell structures during the  
238 hydration process [42-44]. Therefore, the aforementioned nano-inclusion enrichment  
239 layer can provide a large number of nucleation sites near the old concrete surface,  
240 thereby compacting the microstructures and further enhancing the physical bonding of  
241 new-to-old concrete interface. The reinforcing effect of the physical bonding between  
242 nano-inclusions reinforced concrete and old concrete is shown in Figure 10.

243

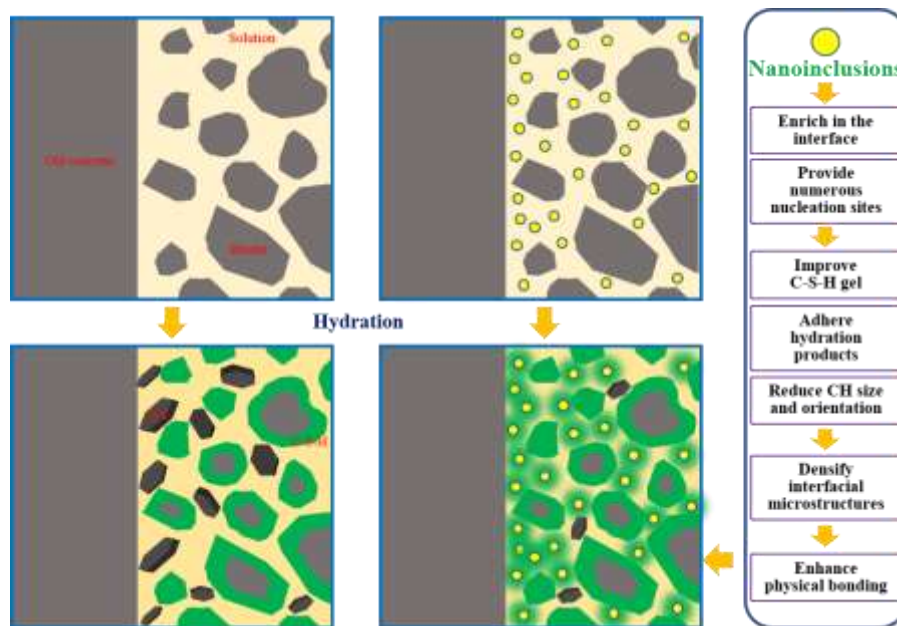


Figure 10. The enhancing effect of physical bonding between nano-inclusions reinforced concrete and old concrete

244

245 The nano-inclusions also have a strong effect on the mechanical interlock between  
246 nano-inclusions reinforced concrete and old concrete. For the polished old concrete  
247 surface, the mechanical interlock is weak because the roughness is low and the binder  
248 particles with the average particle size of 30  $\mu\text{m}$  cannot be hydrated in capillary pores  
249 with the pore diameter of tens of nanometers [45]. In contrast, as shown in Figure 11,

250 the nano inclusions migrate into the micropores of the old concrete with water and  
251 provide nucleation sites at the early stage, allowing hydration products in micropores  
252 connect with that in bulk new concrete, thus enhancing the mechanical interlock  
253 between nano inclusions reinforced concrete and old concrete.  
254

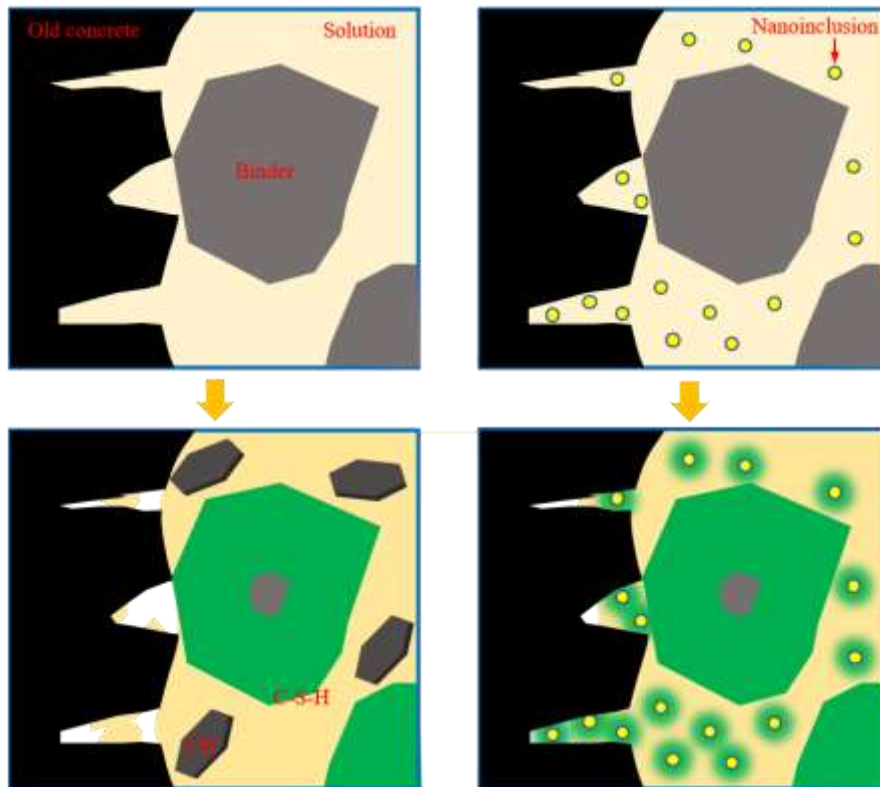


Figure 11. The enhancing effect of mechanical interlock between nano inclusions reinforced concrete and old concrete

### 255 3.3 Prediction model of bond strength

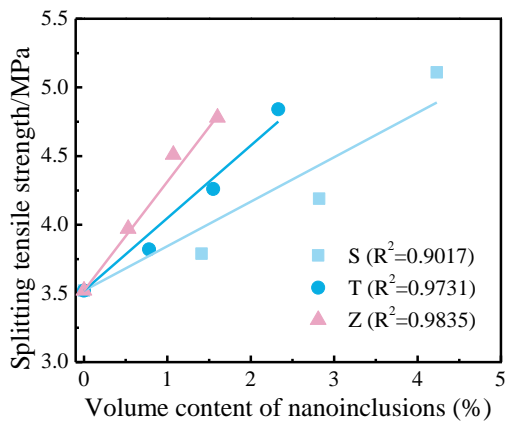
256 Based on the previous analysis, the bond strength between nano inclusions reinforced  
257 concrete and old strength is higher than that between new concrete without  
258 nano inclusions and old concrete due to the enhancement of physical bonding and  
259 mechanical interlock. The physical bonding, mainly van der Waals forces, is affected  
260 by the microstructures of concrete. Therefore, this enhancement is based on the effect  
261 of nano inclusions on the microstructures of concrete, represented by the splitting tensile  
262 strength of nano inclusions reinforced concrete. The splitting tensile strength of

263 nanoinclusions reinforced concrete can be predicted by Equation (3).

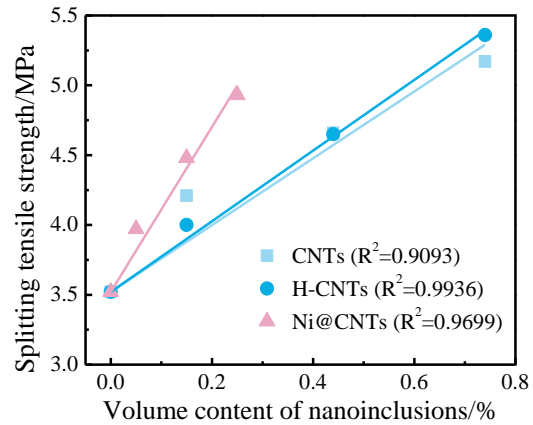
$$f_{tn} = f_t(1 + \alpha V) \quad (3)$$

264 where  $f_{tn}$  is the splitting tensile strength of nanoinclusions reinforced concrete,  $f_t$  is  
 265 the splitting tensile strength of concrete without nanoinclusions,  $V$  represents the  
 266 volume content of nanoinclusions in the concrete,  $\alpha$  is the nanoinclusion enhancement  
 267 coefficient, which can be fitted by experimental results. The fitting results of the  
 268 splitting strength of nanoinclusions reinforced concrete are illustrated in Figure 12 and  
 269 Table 4. It can be seen that the fitting degree of the relation between nanoinclusion  
 270 volume content and splitting tensile strength of nanoinclusions reinforced concrete is  
 271 high; the goodness of fit ( $R^2$ ) ranges from 0.839-0.9936. Based on the fitting results, it  
 272 can be deduced that the enhancement effect of nanoinclusions on the splitting tensile  
 273 strength of concrete can be generally reflected by the geometrical size, i.e. nanosheets >  
 274 nanotubes > nanoparticles.

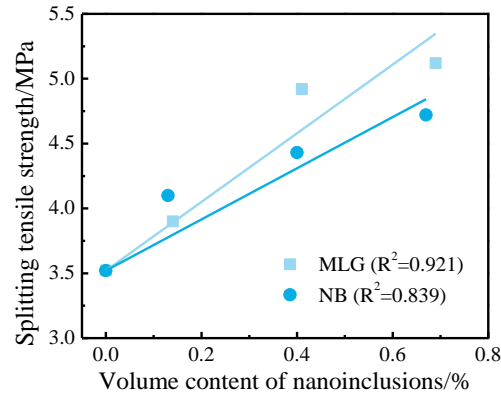
275



(a) Fitting results of splitting tensile strength of concrete with nanoparticles



(b) Fitting results of splitting tensile strength of concrete with nanotubes



(c) Fitting results of splitting tensile strength of concrete with nanosheets

Figure 12. Fitting results of splitting tensile strength of concrete with nano-inclusions

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277

In addition, the enhancement of bond strength of nano-inclusions reinforced concrete

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and old strength is also attributed to the mechanical interlock. As aforementioned, the

279

nano-inclusions can enter the micropores on the old concrete surface and provide

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nucleation sites, thus increasing the mechanical interlock between nano-inclusions

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reinforced concrete and old concrete. It can be deduced that this enhancement is highly

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related to the viscosity of fresh concrete with nano-inclusions, highly consistent with the

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previous research results [8, 46]. The content of nano-inclusions in the micropores of

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the old concrete surface is determined by the total flow of the solution migrating into

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the micropores. For fresh concrete, the liquid flow rate  $Q$  in the micropores can be

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written as Equation (4) [47].

$$Q = - \left( \frac{1}{8\eta} \int_0^\infty r_i^2 \Omega_i dr_i \right) \frac{dP}{dx} \quad (4)$$

287

where  $\eta$  is fluid viscosity;  $r_i^2 \Omega_i$  is the intrinsic properties of micropores on the old

288

concrete surface, in which  $r_i$  is pore radius and  $\Omega_i$  is the average area distribution

289

function of pore  $i$  exposed on any arbitrary face cut perpendicular to the flow;  $dP/dx$

290

is fluid pressure gradient. Therefore, the ratio of liquid flow of fresh concrete with

291

nano-inclusions to that without nano-inclusions can be calculated from Equation (5).

$$\frac{Q_n}{Q_0} = \frac{-\left(\frac{1}{8\eta_n} \int_0^\infty r_i^2 \Omega_i dr_i\right) \frac{dP}{dx}}{-\left(\frac{1}{8\eta_0} \int_0^\infty r_i^2 \Omega_i dr_i\right) \frac{dP}{dx}} = \frac{\eta_0}{\eta_1} = \frac{1}{\eta_r} \quad (5)$$

292 where  $Q_n$  and  $Q_0$  is the liquid flow rate of fresh concrete with and without  
 293 nanoinclusions, respectively;  $\eta_n$  and  $\eta_0$  is the plastic viscosity of fresh concrete with  
 294 and without nanoinclusions, respectively;  $\eta_r$  is the relative plastic viscosity.  
 295 According to reference [48-51], the plastic viscosity of fresh concrete can be predicted  
 296 based on the cell method, as Equations (6)-(10).

$$\eta_r = 1 + \eta_i \lambda \quad (6)$$

$$\lambda = y^3 \frac{4(1 - y^7)}{4(1 + y^{10}) - 25y^3(1 + y^4) + 42y^5} \quad (7)$$

$$y = (\varphi/\varphi_{max})^{1/3}(1 - K) \quad (8)$$

$$K = 3.8 \cdot \frac{V_{sp}}{V_c} \cdot \frac{V_w}{V_s} \quad (9)$$

$$\varphi_{max} = 1 - 0.45 \cdot \left(\frac{D_{10}}{D_{90}}\right)^{0.19} \quad (10)$$

297 where  $\eta_i$  is the intrinsic viscosity;  $\varphi$  is the volume concentration of solid particles in  
 298 fresh concrete,  $\varphi_{max}$  is the maximum packing density;  $V_{sp}$ ,  $V_c$ ,  $V_w$ ,  $V_s$  is the volume  
 299 concentration of superplasticizer, cement, water, solid particle in fresh concrete,  
 300 respectively;  $D_{10}$  and  $D_{90}$  is the sieve sizes corresponding to 10% and 90%,  
 301 respectively.

302 As for the prediction of the bond strength of nanoinclusions reinforced concrete and  
 303 old concrete, the bond strength can be written as in Equation (11).

$$f_b = \beta f_{to} \quad (11)$$

304 where  $f_b$  is the new-to-old concrete bond strength;  $\beta$  is the interfacial strength  
 305 coefficient that equals to the ratio of the bond strength to the splitting tensile strength  
 306 of old concrete;  $f_{to}$  is the splitting tensile strength of old concrete. The incorporation

307 of nano-inclusions can enlarge the interfacial strength coefficient by compacting the  
308 microstructures, but undermine it due to the liquidity loss of fresh concrete. **Therefore,**  
309 **the interfacial bond strength coefficient of nano-inclusions reinforced concrete and old**  
310 **concrete can be written as Equation (12).**

$$\beta_n = \beta_0 \left( 1 + \frac{\alpha\phi}{\eta_r} V \right) \quad (12)$$

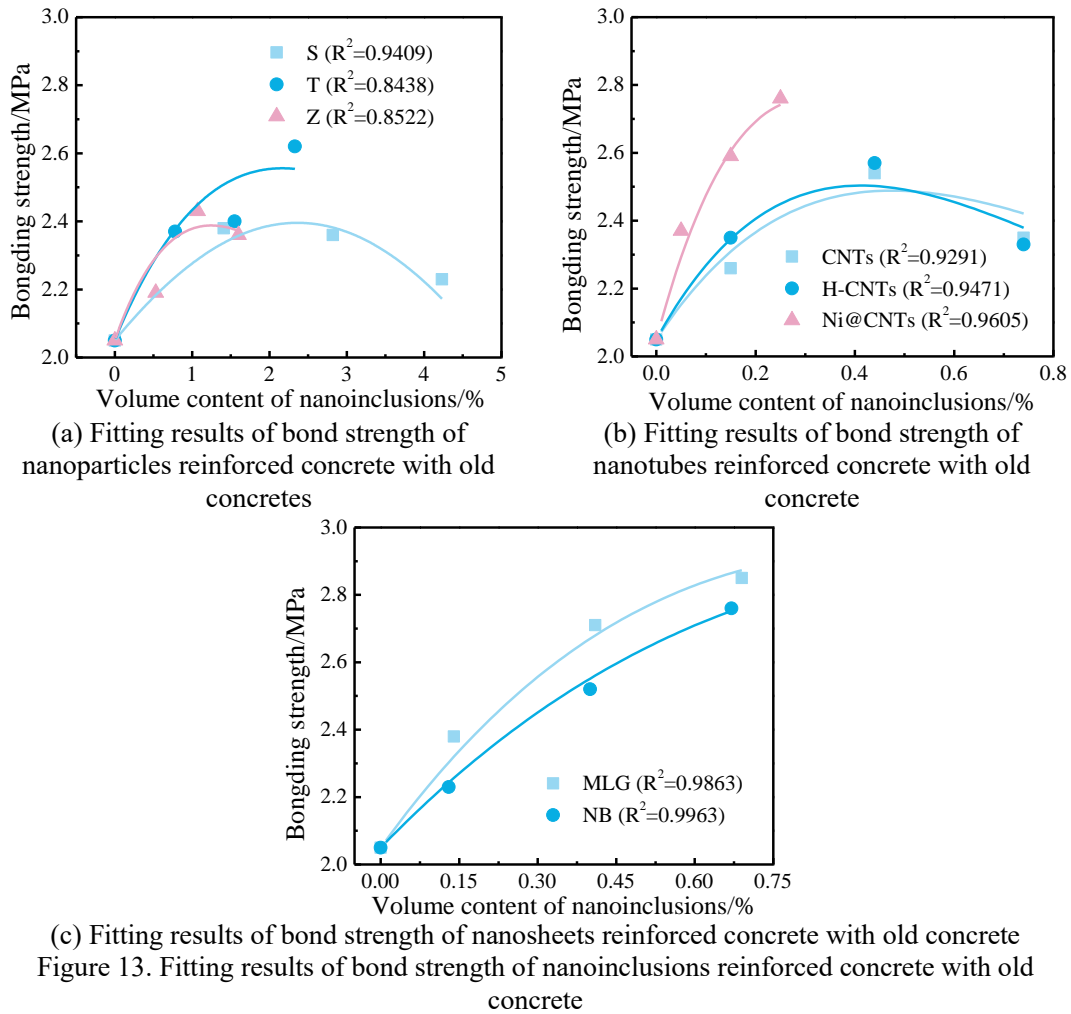
311 where  $\beta_n$  is the interfacial strength coefficient of nano-inclusions reinforced concrete  
312 with old concrete;  $\beta_0$  is the interfacial strength coefficient of new concrete without  
313 nano-inclusions with old concrete;  $\phi$  is the nano-inclusion enrichment coefficient. In  
314 this experiment, superplasticizer is used as the dispersant of nano-inclusions, therefore  
315 part of superplasticizer is adsorbed by nano-inclusions, which reduces the plastic  
316 viscosity of fresh concrete. Therefore, the effective volume concentration of  
317 superplasticizer  $V_{sp,eff}$  can be written as in Equation (13).

$$V_{sp,eff} = V_{sp} \cdot (1 - \gamma V) \quad (13)$$

318 where  $\gamma$  is the weakening coefficient of nano-inclusions to superplasticizer. Combining  
319 Formula (6)-(13) and experimental results, the  $\gamma$  can be fitted, as shown in Figure 13  
320 and Table 4. It can be seen that the fitting degree of the relation between nano-inclusion  
321 volume content and bond strength of nano-inclusions reinforced concrete with old  
322 concrete is high; the  $R^2$  ranges from 0.8438-0.9963. In addition, the fitted nano-inclusion  
323 enrichment coefficients of T, Z, and NB is highly consistent with the experimental  
324 results in this study. Based on the fitting results, it can be concluded that the proposed  
325 prediction model can accurately describe the relationship of the nano-inclusion content  
326 and the bond strength of nano-inclusions reinforced concrete with old concrete.

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Table 4. Fitting parameters of bond strength of nano-inclusions reinforced concrete with old concrete

Nano-inclusion	Nano-inclusion enhancement coefficient $\alpha$	Nano-inclusion enrichment coefficient $\phi$	Weakening coefficient $\gamma$
S	9.188	1.8	14.3162
T	15.029	2.0	37.4400
Z	22.577	1.4	58.3822
CNTs	67.962	1.6	160.8399
H-CNTs	71.866	1.8	177.2741
Ni@CNTs	167.96	1.6	234.8379
MLG	75.232	1.4	74.2485
NB	56.067	1.4	56.6331

331

## 332 4 Conclusions

333 The bond strength and its reinforcing mechanisms and prediction model of  
334 nano-inclusions reinforced concrete with old concrete have been investigated in this  
335 study. The following conclusions can be drawn:

336 • The bond strength between nanoinclusions reinforced concrete and old  
337 concrete is higher than that between new concrete without nanoinclusions and old  
338 concrete.

339 • The mechanisms for bond strength enhancement of nanoinclusions reinforced  
340 concrete with old concrete can be attributed to the improvement of the interfacial  
341 microstructures owing to the enriched presence of nanoinclusions in the new-to-old  
342 concrete interface. The enriched nanoinclusions can modify the compactness of the  
343 hydration products in the interface. The compact microstructures contribute to enhance  
344 the van der Waals forces between nanoinclusions reinforced concrete and old concrete  
345 as well as allow hydration products in micropores with that in bulk new concrete, thus  
346 enhancing the mechanical interlock between nanoinclusions reinforced concrete and  
347 old concrete.

348 • The proposed prediction model can accurately describe the relationship of the  
349 nanoinclusion content and the bond strength between nanoinclusions reinforced  
350 concrete and old concrete. This model can be used to guide the application of  
351 nanoinclusions reinforced concrete in the field of concrete repair.

352 In summary, this study demonstrates that the nanoinclusions reinforced concrete  
353 is a promising material for concrete rehabilitation and strengthening due to its reliable  
354 bonding with old concrete. Moreover, the mechanisms and model proposed in this paper  
355 provide references for further research and a basis for controlling the repair effects of  
356 nanoinclusions reinforced concrete.

357

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362 Conflict of Interest: The authors declare that they have no conflict of interest.

363

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