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Tea stain-inspired treatment for fine recycled concrete aggregates

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HIGHLIGHTS

- Naturally occurring compound, Tannic acid is used to treat fine recycled concrete aggregate (RCA).
- Microstructure and quality of fine RCA are significantly improved by the treatment.
- Over 50% reduction on the total porosity of the mortar can be achieved by the treatment.
- Up to 25% increase in compressive strength can be achieved by the proposed treatment.

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ABSTRACT

A massive fraction of recycled concrete aggregates (RCAs) is fine RCA. Once recycled in new concrete, fine RCA can significantly reduce the mechanical strength and durability of the concrete since fine RCA has high water absorption and high porosity. To address this issue, this study proposes to treat the fine RCA using a tea-stain inspired compound, tannic acid (TA). To this end, fine RCA is first immersed in dilute solution of TA. In this process, TA can react with calcium hydroxide (CH) and calcite of RCA to produce nanoparticles on the surface of the RCA particles. These nanoparticles can fill the capillary nanopores in the mortar, significantly reduce the porosity of the produced mortar. This can be confirmed by the scanning electron microscopy (SEM) and mercury intrusion porosimetry (MIP) analysis. Experimental studies have been carried out to examine the effect of this treatment on the hydration and mechanical behavior of the mortars made with the treated fine RCA. Testing results confirm that the strength and durability of the mortar can be significant enhanced by the proposed method, which can be mainly attributed to two reasons. First, TA coated on the surface of the fine RCA can capture calcium ions to induce local mineralization and facilitate the hydration of cement in the interface transition zone (ITZ), densifying the ITZ according to nanoindentation test. Second, the cohesion of hardened cementitious material can be enhanced by the unique binding capacity of TA through covalent and non-covalent interactions.

1. Introduction

Construction and demolition waste (C&D waste) is one of the largest solid wastes in many countries, especially in China [1–3]. Most of them are disposed of in landfills, which not only consumes enormous space for landfills, but also creates various pollutions in land, water and air [1,4]. Recycling C&D wastes for new applications is the most effective way to alleviate the problems created by C&D wastes. For example, end-of-life concrete, which is the heaviest C&D waste, can be crushed into suitable particles size to be used as coarse or fine aggregates in new concrete [1,5–12]. The crushed particles are usually classified as coarse recycled aggregates (RCAs) if their size is no less than 4.75 mm and fine RCAs if their size is less than 4.75 mm.

Numerous studies have shown that the coarse RCAs are highly porous due to the residual cement mortar attached to the original aggregates [1,5–9]. As a result, coarse RCAs have inferior properties such as higher water absorption, higher porosity, lower particle density and higher crushing value than natural aggregates (NAs) [7,10–12]. Consequently, the performance of the concrete with coarse RCAs is inferior to the concrete with NAs [1,6,7,13,14]. Many methods have been developed to improve the performance of coarse RCA, through either removing [8,15,16] or strengthening the adhered cement mortar [10,17,18].

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Up to now, study on treating fine RCA is very little, despite large amount of fine RCA is produced in crushing and screening processes. Fine RCA is actually discouraged to be used in concrete due to its high water absorption, high porosity, poor microstructure, especially at the interface transition zone (ITZ) between the new cement matrix and the fine RCA [15]. The performances of concrete (including workability, strength and dry shrinkage) made with fine RCAs are inferior to those of the concrete made with NAs [4,16,17]. For example, Khatib found that strength is reduced by 15–30% for concrete containing fine RCA [18]. Bravo et al. [19] compared concrete samples made with fine RCAs from three different plants and found their compressive strengths at 28d are 18–42% lower than that of the control one. Zhao et al. [20] demonstrated that the compressive strength of mortars decreases quasi linearly as the replacement percentage of fine RCA increases.

A few treatment methods have been proposed to improve the quality of fine RCA. For example, Zhang et al. [21] attempted to strengthen the adhered mortar through carbonating the fine RCA. In their study, the fine RCA was exposed to 20% CO_2 concentration with 60% relative humidity in a chamber for 7d. After treatment, water adsorption of treated fine RCA was decreased by 28.3%, and crushing value was decreased by 9.1% compared with non-carbonated fine RCA. Shi et al. [22] exposed fine RCA to the 20% CO_2 concentration with 60% relative humidity in a chamber for 3d. They found that water adsorption of the fine RCA was reduced by 35.9%, and apparent density was increased by 2.9% by the carbonation. As a result, the compressive strength and durability of mortar with the carbonated fine RCA were significantly improved. Guo et al. [23] presented the physical strengthening treatment of particle through removing the adhered mortar. Their result showed that the crushing value of fine RCA was reduced from 22% to 17% after the treatment. The separated fine powder with 0.16 mm can be recycled as active powder used in partial replacement of cement. The fine powder can be further improved by heating process [24]. Fine RCA was also treated by enhancing the interfacial transition zone (ITZ), which can be done by spraying polymer emulsion or immersing fine RCA in sodium silicate solution or pozzolan slurry, or depositing calcium carbonate through the metabolism of bacteria [22,25–28].

In this study, we propose a new method to treat fine RCA with tannic acid (TA) so they can be used in new concrete with improved mechanical properties. This idea is inspired by the daily life observation which shows that tea always leaves stain on the glassware. This suggests that some compound in the tea has strong bonding ability. Inspired by this observation, early civilizations were able to cure leather products with tea extract. The material responsible for the tea staining and tanning has been identified as TA [29,30], which is a plant polyphenol, the world's third largest class of plant components [31]. One unique feature for this material is its capability to strongly bind to diverse surfaces by covalent interaction and non-covalent interactions [32]. A recent study by Oh et al. [33] demonstrated that TA has ability to capture calcium ions and induce local mineralization of hydroxyapatite (HA). This is because the pyrogallol group of the TA can bind Ca^{2+} in aqueous environments [34], which facilitates a local super-saturation of Ca^{2+} to induce faster localized mineralization of HA. Hydration of ordinary Portland cement (OPC) is nothing but a mineralization process of hydration products such as calcium silicate hydrate and calcium hydroxide (CH). It is reasonable to speculate that TA can facilitate the hydration of OPC in the similar fashion as it does on the mineralization of HA shown in [33,34]. Therefore, a tea-stain-inspired method is proposed to treat fine RCAs. In this method, the fine RCA is simply immersed in a diluted TA solution before added into the concrete mix. It is expected a few mechanisms can be introduced by this treatment. First, chemical reaction between TA and hydration products such as calcium hydroxide can occur which densifies the microstructure of the fine RCA. Second, excellent bonding ability of TA coated on the fine RCA can enhance the cohesion of the produced mortar. Third, TA may facilitate the hydration of cement near the interface between the new cement paste and the RCA so that a denser microstructure can be created. As a result, the performance of concrete/mortar with the fine RCAs can be greatly improved by this treatment.

2. Materials and methods

2.1. Materials

Waste concrete cylinders were collected from a laboratory at the University of Alabama. The waste concrete was crushed into particles by a jaw crusher (manufactured by BICO Inc.) which were sieved into coarse and fine RCAs. The fine RCA was used to manufacture cement mortars with OPC. Fig. 1 shows the particle size distributions of this aggregate. Its water absorption is determined as 11.3%. Table 1 shows the chemical components as oxide form of the OPC. River sand was chosen as the natural fine aggregate to produce control mortar samples. TA is analytical reagent with over 95% purity.

2.2. Treating and characterizing the fine RCA

The fine RCA was dried in an air-drying oven to remove the freedom water before soaking in the TA solution. TA solutions with five mass concentrations (0.1%, 0.3%, 0.5%, 0.75% and 1%) were prepared before the treatment. These solutions were made by simply dissolving the calculated amount of TA powder into tap water. Then the dried fine RCA was immersed in these TA solutions at solid to liquid ratio of 1.5 kg/L at 23 °C ± 2 °C. The pH values of all TA solutions were recorded during soaking process. After 2 h soaking, fine RCA was filtered out and soaked in fresh water for another 2 h to remove the residual TA. After that, the treated fine RCA was filtered out and dried in the air-drying oven at 70 °C until the its mass reached a constant.

Water absorption of fine RCA was measured according to ASTM C 128-15 [35], which is the same as Chinese standard GB/T 14684-2011 [36]. The test was carried out as follow:

1) The fine RCA sample was immerse in fresh water for 24 h at room temperature 23 °C. Then, the sample was dried with an air blower to remove the surface moisture until the sample reached the saturated surface dry state. The mass of sample at saturated surface dry state was recorded as m_s.

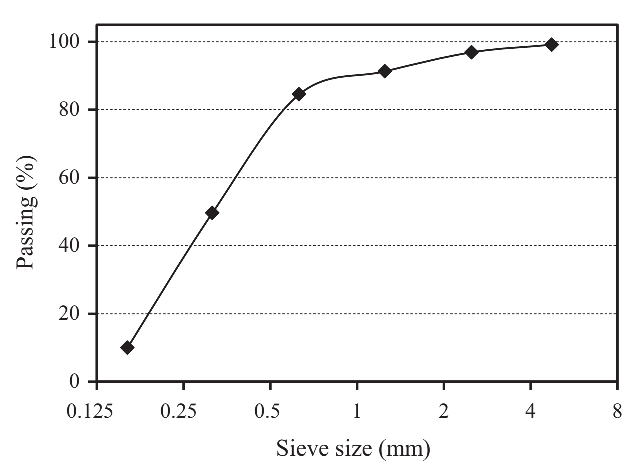

Fig. 1. Particle size distribution of the fine RCA.

Table 1
Chemical compositions of the OPC.

Compositions	CaO	SiO_2	Al_2O_3	Fe_2O_3	MgO	K_2O	Ti_2O	P_2O_5	Na_2O	LOI
Percentage (%)	64.92	21.15	5.92	3.15	0.83	0.57	0.26	0.19	0.10	2.58

Table 2
Mix proportions (kg/m^3) of mortar with the pristine and treated fine RCA.

Constituents	Water	Cement	Fine aggregate	Superplasticizer
Mortar with fine RCA	274	457	1469	1.37
Mortar with river sand	217	457	1469	1.37

2) the sample was dried in an oven with temperature of 105 °C for at least 24 h until the mass of the sample became a constant. The mass of this sample was recorded as the dry mass m_d. The water absorption (W_a) of the fine RCA was calculated by the follow equation.

$$W_a = (m_s - m_d)/m_d \quad (1)$$

The mineral compositions produced by the TA treatment was analyzed by X-ray diffractometer (XRD). Fourier Transform Infrared (FTIR) Spectrometer was used to examine the chemical bonds of pristine and treated fine RCAs. Scanning electron microscopy (SEM) was used to study the microstructure difference between the pristine and treated fine RCAs. The change of pore structure of the fine RCA induced by the treatment was examined by a fully automatic physical adsorption instrument. To this end, fine RCA was first ground into particles smaller than 165 μm. The pore size distribution of these particles was then determined by Brunauer-Emmett-Teller (BET) theory using nitrogen adsorption.

2.3. Manufacturing and characterizing the mortar specimens with RCAs as fine aggregates

2.3.1. Manufacturing process of mortar specimens

Mix design of mortars made with pristine and treated fine RCAs is detailed in Table 2. Same water/cement ratio (0.60) was used to produce all mortar samples. The superplasticizer was added in the mixture to increase the workability of the mortar. Both the pristine and treated RCAs were used to manufacture the mortar samples. As a comparison, a group of mortars (noted as "NA") was made with natural fine aggregate (river sand). The mortar with NA was manufactured by replacing all fine RCA with river sand and adjusting the water content to ensure the same amount of free moisture in this mortar as in those with RCA. The fine RCA has a 11.3% water absorption and 6.5% water content when added into the mortar; while the water absorption of NA is only 0.9%. Therefore, the amount of water used for the mortar with NA is reduced by 57Kg/m^3 to reach the same amount of free moisture as those of the mortar with fine RCA, as shown in Table 2.

The fresh mortars were cast into 50 mm × 100 mm cylinder molds and 40 mm × 40 mm × 160 mm prism molds. The cylinder specimens were used to measure the compressive strength, and the prism specimens were used to measure the flexural strength. All fresh specimens were cured in standard curing room at temperature of 23 °C ± 2 °C and relative humidity above 95% until the testing age. The compressive strength test was carried out at the ages of 3 d, 7 d, 28 d and 90 d, and the flexural strength test was carried out at the ages of 28 d and 90 d.

2.3.2. Characterizing the mortar specimens

Flowability of mortars was measured to evaluate the workability of mortars according to ASTM C1437 [37]. Isothermal calorimetry test was carried out to study the effect of the treatment on the cement hydration according to ASTM C1679 [38] and ASTM C1702 [39] using a I-Cal 2000HPC (Calmetrix Incorporation).

The porosities of the produced mortars were further examined by Mercury intrusion porosimetry (MIP). Nanoindentation was carried out to characterize nanomechanical properties of the ITZ between new and old matrix of the mortar samples using an Agilent Technologies G200 Nanoindenter. Specimens were prepared in consistent with previous nanoindentation studies [40]. Various arrays of the indent tests were performed to probe the microstructural properties across the selected region. The nanoindentation tests were performed in load control mode with maximum load of 0.7 mN in order to achieve acceptable depth range [41]. The maximum load was attained through five cycles of a trapezoidal load history where the time to load, hold, and unload were all 10 s. Progressively loading each indent in cycles allows for easier identification of inhomogeneity in the indentation volume, although it is more time consuming. The nanoindentation modulus was calculated for the last cycle of each test using the Oliver and Pharr method [42] and the calculation is described fully in elsewhere [43,44].

3. Results and discussions

Fig. 2 compares the pristine fine RCA and the one treated in 0.5% TA solution. The treated fine RCA exhibits a brown color in comparison with the grey color of the pristine one due to the precipitate deposited on the surface of the fine RCAs produced by the TA treatment.

3.1. pH values of the TA solutions

Fig. 3 presents pH values of all treating solutions varying with the soaking time. In this figure, "Control" refers to the pH value

Fig. 2. Specimens of pristine and treated fine RCAs: (a) pristine sample; (b) treated with 0.5% TA.

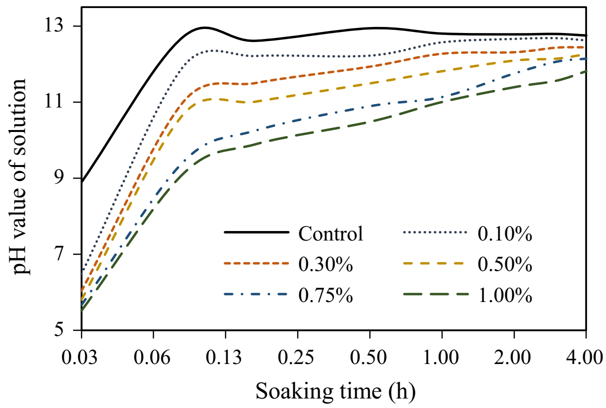

Fig. 3. pH values of the TA solutions varying of soaking time.

of the water without TA; "0.1%", "0.3%", "0.5%", "0.75%" and "1%" refer to the mass concentrations of the TA solution used to treat the fine RCA. Overall, the solution with higher TA concentration has a lower pH value and the pH values of all TA solutions are lower than 7. This acidic nature of all these solutions changes to basic after the fine RCA was added, as shown in Fig. 3. The pH values of all TA solutions increase with soaking time, especially first 10 min, due to the dissolution of portlandite (CH) from the fine RCA, which reacted with TA. After 2 h, pH values of these solutions increase very little with time, suggesting that most chemical reaction between TA and RCAs is completed within 2 h. For this reason, 2 h soaking was used in all experiments in this study.

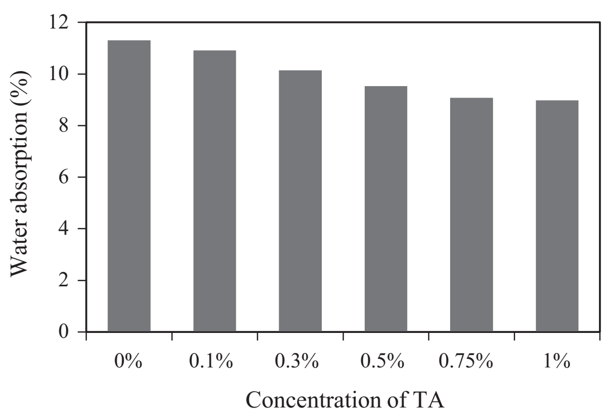

Fig. 4. Water absorption of the pristine and TA treated fine RCA.

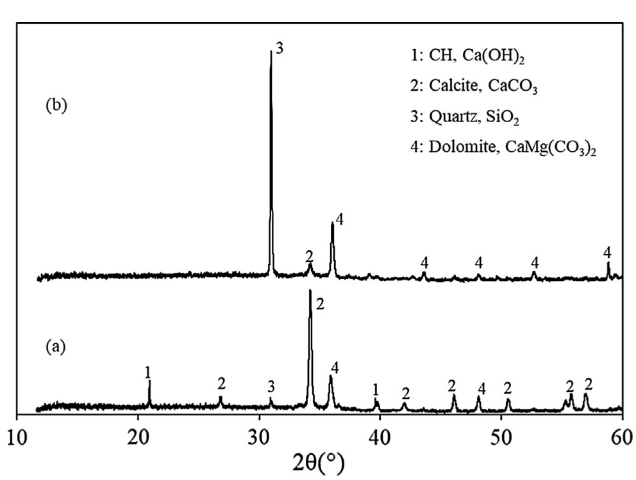

Fig. 5. XRD patterns of pristine and treated fine RCA: (a) pristine; (b) treated with 0.5%TA.

3.2. Effects of TA treatment on the properties of fine RCAs

3.2.1. Water absorption

Fig. 4 presents the effect of TA treatment on water absorption of the pristine and treated fine RCAs. It can be seen that the pristine RCA has a very high water absorption. It can be reduced by the proposed treatment. Increasing the concentration of TA solution, more reduction in water absorption can be achieved, as revealed by Fig. 4.

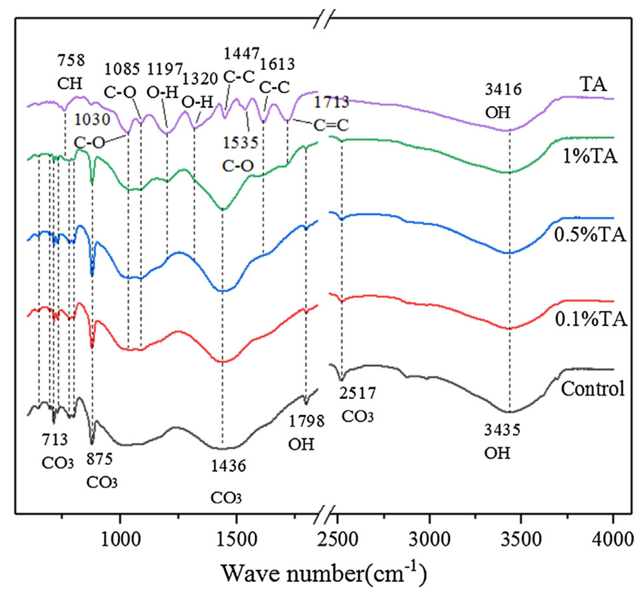

Fig. 6. FTIR analysis of pristine and treated fine RCA.

Table 3
FTIR properties of TA, pristine and TA treated fine RCA (wave number: cm^{-1}).

RCA	0.1%TA	0.5%TA	1% TA	TA	Assignment
3435	3427	3427	3416	3416	Stretch vibrations of O–H in H_2O
2517	2520	2520	2520	–	Out of plane bend and split in plane of bending vibration of CO_3 in calcite
1798	1798	1797	1795	–	(–OH) stretch of $Ca(OH)_2$
–	–	–	1701	1713	(C = C) stretch of benzene ring
–	–	1595	1594	1613	(C–C) stretch and (C–H) deformation in plane of benzene ring
–	–	–	–	1535	(C = O) stretch of carboxylic acid
1436	1436	1436	1440	1447	Asymmetric stretch of CO_3 in calcite and C–C stretch of benzene ring
–	–	–	–	1320	(–OH) stretch in plane of benzene ring
–	–	1163	1200	1197	(O–H) deformation in plane of phenolic and carboxylic acid, (C–C) stretch and (C–H) deformation in plane of benzene ring
–	1085	1084	1081	1085	(C = O) stretch of carboxylic acid
–	1036	1036	1040	1030	(C = O) stretch of carboxylic acid
875	876	876	876	874	split in plane of bending vibrations of CO_3 in calcite
797	797	797	797	–	(Si-O) stretch of quartz
779	779	779	778	–	(Si-O) stretch of quartz
–	–	–	756	758	(C–H) torsion of benzene ring
728	729	729	729	–	(–OH) stretch of $Ca(OH)_2$
713	713	713	713	–	Out of plane bend of CO_3 in calcite
646	646	646	647	–	Si-O stretch of quartz

3.2.2. XRD, FTIR and SEM analysis

XRD patterns of the pristine and the treated fine RCAs are show in Fig. 5. CH, calcite, quartz and dolomite are the main crystal compositions in the pristine fine RCA, as shown in Fig. 5(a). The dolomite comes from the natural coarse aggregates in the waste concrete. The presence of calcite in the pristine fine RCA is induced by the carbonation of CH on the surface of the RCA in the air. After treating with 0.5% TA solution, peaks of CH and some peaks of calcite are absent, as shown in Fig. 5(b), suggesting that CH and some calcite were consumed by the chemical reaction with the TA. As a result, some cement paste was removed so that more natural sand was exposed on the surface of the treated RCA. Consequently, the peak of quartz increased after the treatment as shown in Fig. 5. However, the products of the reaction between the TA and CH couldn't be detected by XRD.

Fig. 6 shows the FTIR spectra results of the TA used for the treatment, the pristine fine RCA (Control), and the treated fine RCAs. In this figure, treated samples are named by the concentration of the TA solution used for the treatment. The positions of peaks and their assignments are detailed in Table 3. The peaks at 713 cm^{-1}, 875 cm^{-1} and 1436 cm^{-1} are caused by out-of-plane bend (ν_2) of CO_3 in calcite, split in-plane of bending vibrations (ν_4) of CO_3 in calcite, asymmetric stretch (ν_3) of CO_3 in calcite, respectively; the peak at 2517 cm^{-1} are caused by out of plane bend and split in-plane of bending vibrations of CO_3 in dolomite; peaks at 728 cm^{-1} and 1798 cm^{-1} are caused by OH stretch of CH. Among those peaks, the characteristic peaks at 713 cm^{-1}, 1798 cm^{-1} and 2517 cm^{-1} are gradually weakened with the increase of TA concentration, suggesting that the calcite and CH were consumed by the reaction between the fine RCA and TA. This is in good agreement with the results of XRD analysis. The (C–C) stretch and (C–H) deformation in plane of benzene ring detected around 1613 cm^{-1} in the TA solution is shifted to the left in the treated fine RCA as shown in Fig. 6, suggesting the formation of TA-Ca complex probably due to the coordination of phenolic-OH group with calcium ions through deprotonation. For this reason, the (C = O) stretch of carboxylic acid around 1030 cm^{-1} and 1085 cm^{-1} was also detected in the samples of the treated fine RCAs. Based on the results of XRD and FTIR analysis, it can be concluded that the calcite and CH in the RCA can react with TA to produce a calcium-TA complex.

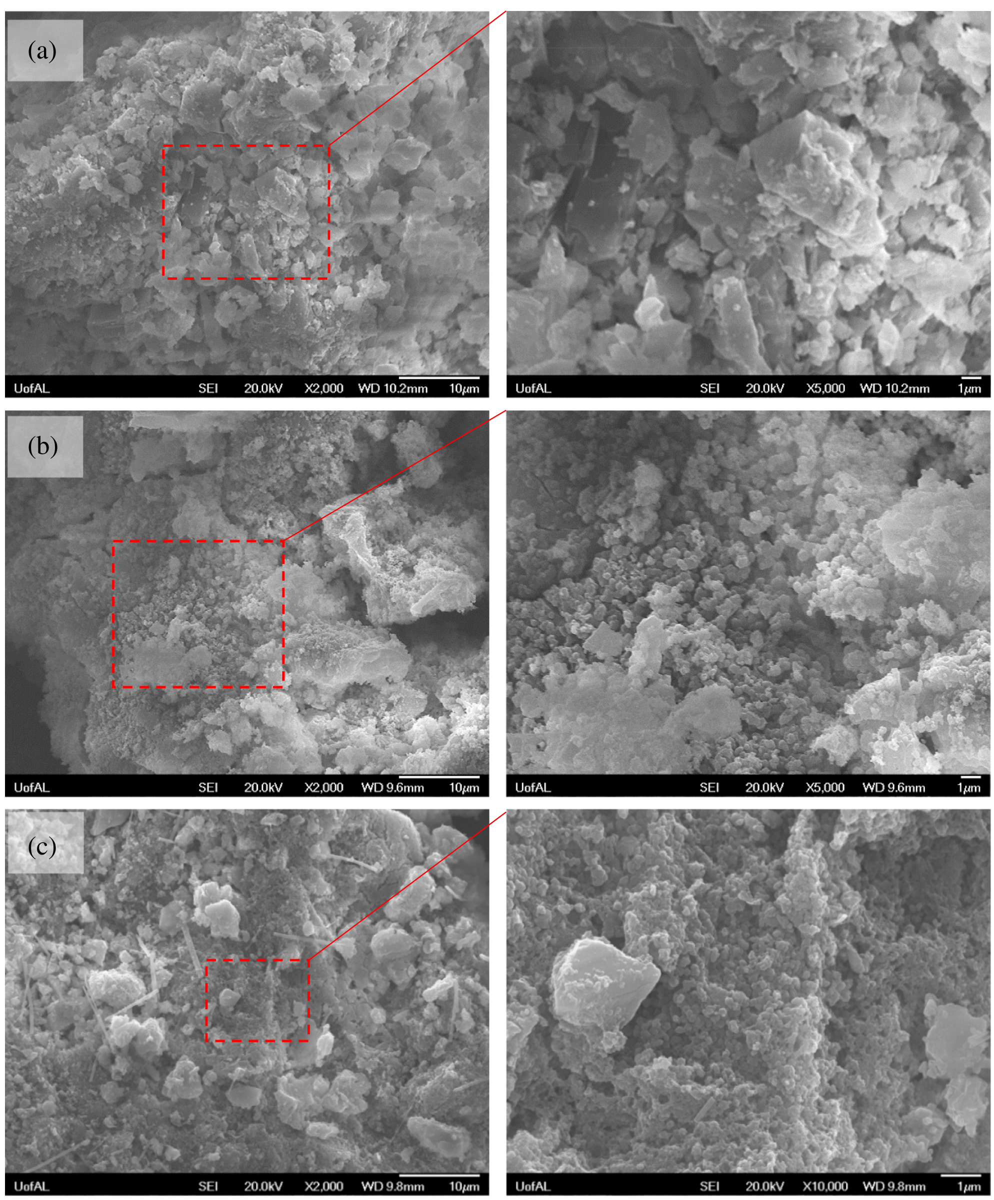

Fig. 7. SEM images of the pristine and the treated RCA: (a) pristine; (b) treated with 0.5% TA; (c) treated with 1% TA.

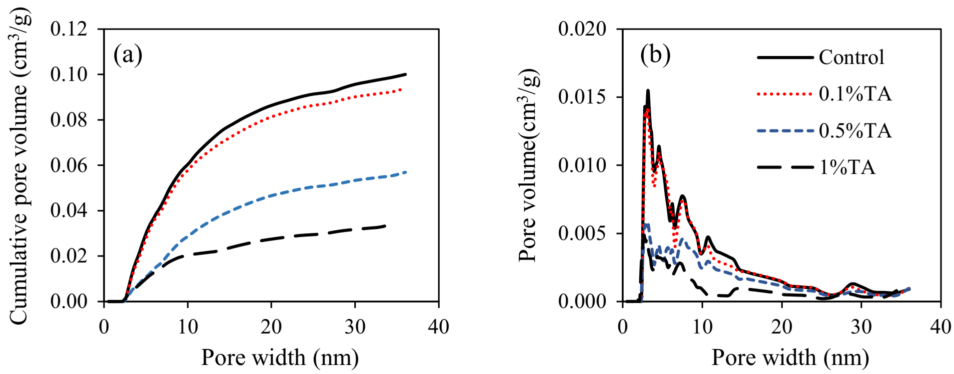

Fig. 8. Effect of TA treatment on the cumulative pore volume and pore volume of the waste concrete powder: (a) cumulative pore volume; (b) pore volume.

Fig. 7 shows the SEM images of the pristine and the treated fine RCA samples. The pristine RCA sample is covered by porous fine particles and loosely bonded cement mortar as observed by Fig. 7(a). Fig. 7(b) and (c) show the surface of the fine RCA treated with 0.5% and 1% TA solutions, respectively. It can be clearly seen that those loosely bonded cement mortar present on the surface of pristine fine RCA was removed after the treatment. More interestingly, large amount of nanoparticles are deposited on the surface RCA, as show in Fig. 7(b) and (c). These nanoparticles were most likely calcium-TA complex, produced by the reaction between CH, calcite and TA, as indicated by XRD and FTIR results shown in Figs. 5 and 6.

Fig. 8 compares the pore structures of the pristine RCA (control) and TA treated RCA powders. As shown in Fig. 8(b), most pores in these small RCA powder samples are less than 40 nm, suggesting they are either small capillary pores or gel pores. These pores can be slightly reduced by treating the RCA with 0.1%TA solution. When higher concentration of TA solution was used, more reduction of the pores can be achieved. Fig. 8(a) shows that the cumulative pore volumes were reduced by 43% and 66% for the RCA treated by TA solutions at 0.5% and 1.0%, respectively. This reduction of pores can be mainly attributed to reaction between the TA and the cement hydration products, which produces many nanoparticles to fill some nanopores, as indicated in Fig. 7. Fig. 8 also explains why the water absorption of the fine RCA can be decreased by the TA treatment.

3.3. Characterization of the mortar with the fine RCAs

3.3.1. Cement hydration

Fig. 9 shows that the cement hydration of mortars with fine RCA in the first 96 h. Two peaks, which are induced by the hydration of C_3S (tricalcium silicate) and C_3A (tricalcium aluminate), respectively, appear for all mortar samples shown in Fig. 9(a). The effect of using fine RCAs treated with 0.1% TA or 0.3% TA solutions on the cement hydration appears negligible. However, adding fine RCAs treated with 0.5% TA solution clearly retards the hydration of the cement, as shown in Fig. 9(a). This retarding effect is induced by the residual TA absorbed by the RCA. Nevertheless, the accumulative hydration heat generated by this sample gradually approaches to the control one with the increase of time as shown in Fig. 9(b). The retarding effect of TA treatment became much more severe when 1% TA was used since more residual TA was available. The arriving time of the first peak was delayed to 38 h and the second peak almost disappeared, indicating that TA is an excellent retarder for cement hydration.

3.3.2. Pore volume and pore size distribution

The effect of the proposed treatment method in pore volume and pore size distributions of a mortar made with the fine RCAs is revealed by Fig. 10. As shown in Fig. 10(a), the total pore volume of the mortar sample was significantly reduced after the TA treatment, which greatly enhances the properties of the fine RCA. Two reasons are responsible for this significant reduction in porosity. First, some loose particles present in the pristine fine RCA were removed after the TA treatment, as indicated by the SEM analysis shown in Fig. 7(b), which can decrease the porosity of the RCA. Second, reaction between CH, $CaCO_3$ and TA produces large amount of nanoparticles deposited on the surface of the TA treated fine RCA, as observed in Fig. 7. Those nanoparticles can fill the pores in the mortar, leading to significant reduction of pores in nanoscale, as shown in Fig. 8. This can be further confirmed by the results of pore size distribution, as observed in Fig. 10(b). Almost half of capillary pores with size between 10 nm and 100 nm were reduced by the TA treatment, which can significantly improve the strength and durability of the mortar. Fig. 10(b) also shows that more reduction on capillary pores with size 10–100 nm can be achieved by treating

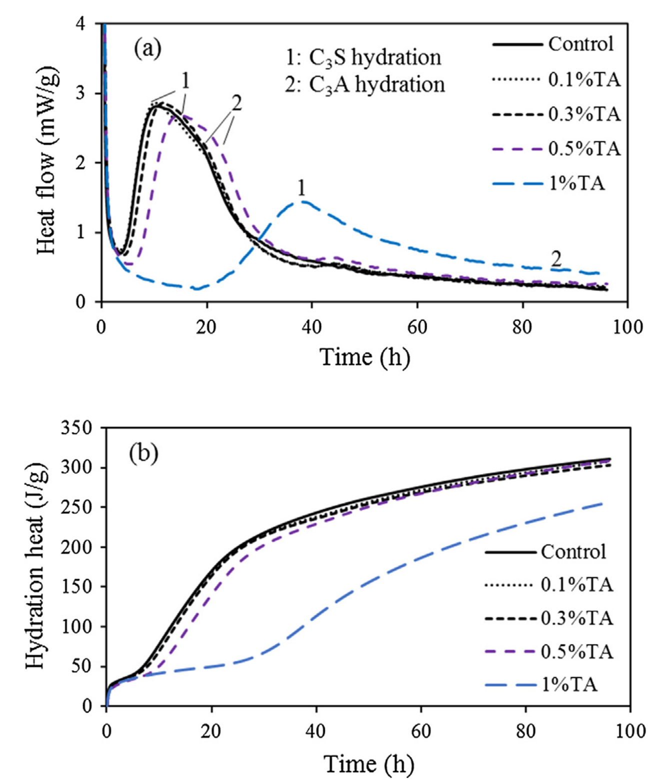

Fig. 9. Effect of the TA concentration on the cement hydration with fine RCA: (a) heat flow of the cement hydration; (b) accumulative heat of the cement hydration.

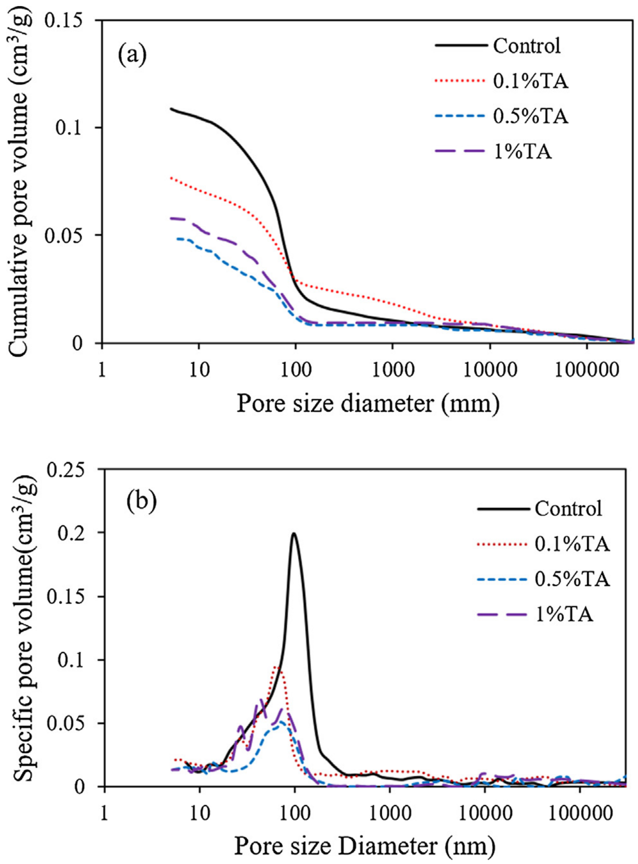

Fig. 10. Effect of TA concentration on the pore volume and pore size distribution of the mortar sample: (a) cumulative pore volume; (b) pore size distribution.

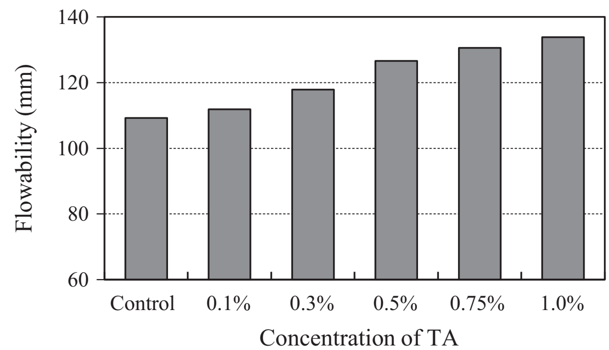

Fig. 11. Effect of TA treatment on the flowability (mm) of mortar with fine RCAs.

the fine RCA with higher concentrated TA solution since more nanoparticles can be deposited on the surface of fine RCA with more TA.

Fig. 10 also indicates there is an optimal content of TA for the RCA treatment. If TA concentration is too low (0.1%), reaction between the TA and RCA is limited, leading to higher porosity of the mortar. However, when 1% of TA solution is used, the hydration of the mortar was significantly retarded, as shown in Fig. 9. As a result, the porosity of the produced mortar became higher than that treated with 0.5% TA solution at the same age.

3.3.3. Mechanical strength

The flowability, compressive strength and flexural strength of the mortar specimens with pristine and treated fine RCAs are presented in Figs. 11–14. As shown in Fig. 11, the flowability of the fresh mortar can be improved by the TA treatment, and the flowability of the mortar increases with the concentration of the TA solution used to treat the fine RCAs. This can be attributed to the lower water absorption, less angular shape and smoother surface of the TA treated fine RCA, as can be observed from Figs. 4 and 7.

Figs. 12 and 13 show the effect of TA treatment on the compressive strength of the mortar. Compared with the mortar with pristine fine RCAs (Control group), all mortar specimens made with TA treated RCA possess higher compressive strength for all ages. This improvement is rather small at the early age because of the retarding effect of TA, as revealed by calorimetry testing (Fig. 9). The retarding effect diminishes at later ages as higher improvement on the compressive strength has been achieved at 28d and 90d, as show in Figs. 12 and 13. The highest increase of the compressive strength was achieved by treating the RCA with 0.75% TA solution. In this case, the compressive strengths of the mortar at 90d is increased by 25%. Figs. 12 and 13 also show that more strength increase can be achieved by using higher concentration TA solution to treat the RCA. This trend holds until the concentration of the TA solution reaches 1%. At such a high concentration, the residual TA in the RCA becomes excessive and its retarding effect significantly reduces the hydration rate of the cement. As a result, the mortar made with 1%TA treated RCA has lower compressive strength than those of other groups. Therefore, no more than 0.75% TA concentration is recommended for the fine RCA treatment to avoid the excessive retarding effect.

Fig. 12 also compares the compressive strengths of the mortars made with fine RCA and the one made with natural fine aggregates (NA group). As expected, the mortar specimen made with pristine fine RCAs is weaker than the NA group at all ages. By using the TA treatment, the compressive strengths of the mortar specimens are much improved and just slightly lower than that of the NA group at 3 d, 7 d, and 28 d. At 90 d, the compressive strengths of mortars made with fine RCA treated by 0.5% and 0.75% TA solution surpass that of the NA group, suggesting the effectiveness of the proposed method on improving the property of fine RCAs and the resulting mortars.

The flexural strengths of the mortars made with fine RCAs at 28d and 90d, respectively, are presented in Fig. 14. It shows that the mortar made with treated fine RCAs has higher flexural strength than the control group. Similar to the case of the compressive strength, the increment on the flexural strength of mortar increases with the age as indicated in Fig. 14. The flexural strength of mortar made with fine RCA treating with 0.5% TA solution is only 0.8% lower than the one with the natural fine aggregate (NA group).

3.3.4. Drying shrinkage of the mortars made with RCA

Fig. 15 compares the drying shrinkage of the mortar specimens made with pristine and treated fine RCAs. Overall, the drying shrinkage of the mortars can be significantly reduced by TA treatment. This significant reduction on drying shrinkage can be attributed to two factors. Frist, TA in the mortar can retain water because of its hydrophilic nature. Second, the porosity of the mortar is lower in the mortars with TA treated RCA. Both factors can slow down the water evaporation from the mortar, leading to lower drying shrinkage of the mortar. Fig. 15 also presents that the fine RCAs treated with higher concentration of TA can reduce more drying shrinkage because more TA is present in the cement mortar. Nevertheless, the drying shrinkages of all mortars with fine RCAs are significantly higher than that of the one with NA after 7 d. At 7 d, only the mortar with 1% TA treated fine RCAs has smaller drying shrinkage than the one made with the natural fine aggregate because of slow cement hydration induced by the 1% TA treatment.

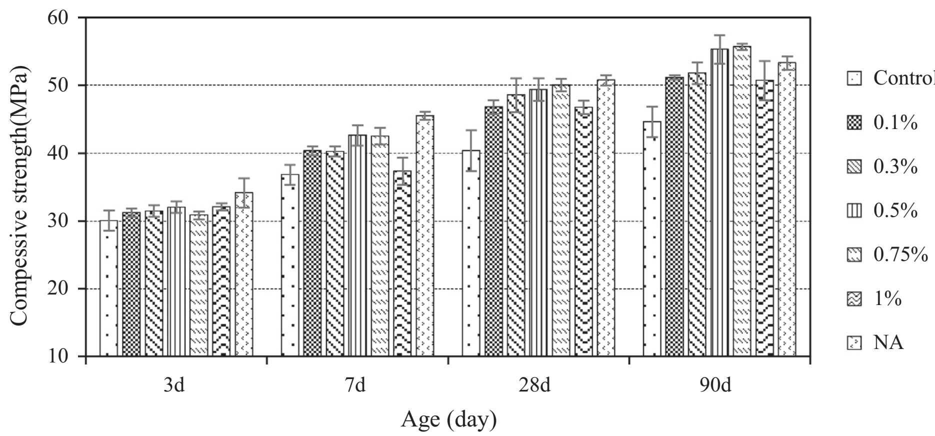

Fig. 12. Compressive strengths of mortar made with pristine and TA treated fine RCAs.

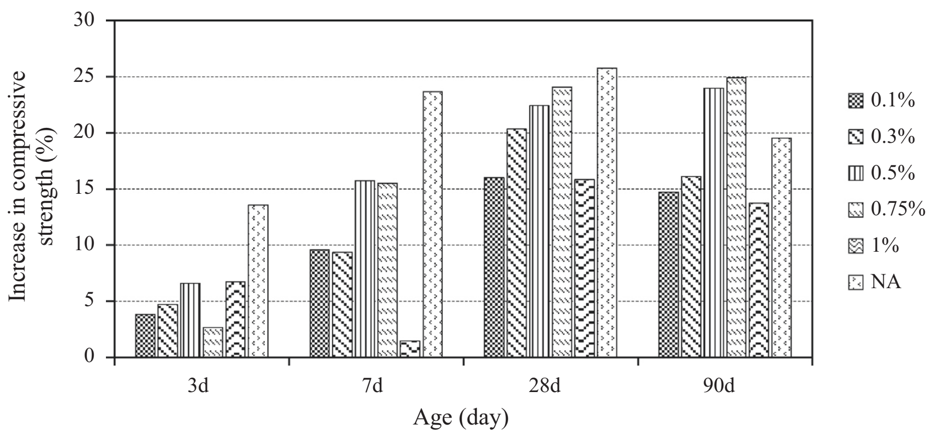

Fig. 13. Compressive strength increment of mortar (%) achieved by treating fine RCAs.

After 7 d, the dry shrinkage of this group rapidly increased and exceeded that of the NA mortar.

The nanoindentation test was carried out to verify whether TA can facilitate cement hydration near the surface of the fine RCA by examining the microstructure and nanomechanical property of the ITZ between the new and old cement matrices. For both ITZs of the control mortar and the one using the TA treated RCA, the indentation were performed on a 10×10 array with 10 µm spacing in both *x* and *y* directions. The polishing procedure was effective enough that there were no aberrant tests found in the reported arrays of tests. The elastic modulus calculated for each test was analyzed spatially using contour mapping [45].

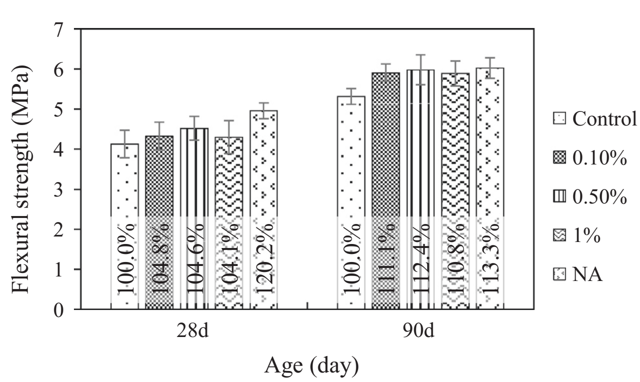

Fig. 14. Flexural strengths of the mortars made with pristine and treated fine RCAs.

Figs. 16 and 17 compare the elastic modulus contours in the ITZ in the mortars made with pristine and TA-treated RCA. In these two figures, some particles with high elastic modulus can be identified, which are sand or unhydrated cement particles. Other area with relatively uniform elastic modulus is the hydration products of cement. In this area, the mortar made with the RCA treated with 0.5% TA has higher indentation modulus than the one made with the pristine fine RCA. This indicates that a dense microstructure was produced in this zone due to the TA treatment, suggesting that

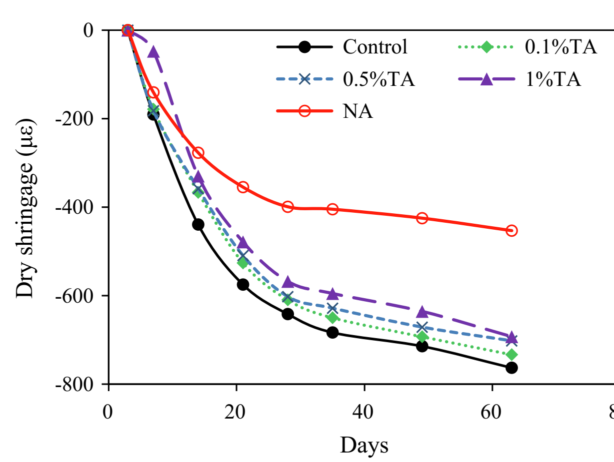

Fig. 15. Drying shrinkage of the mortars with pristine and TA treated RCA.

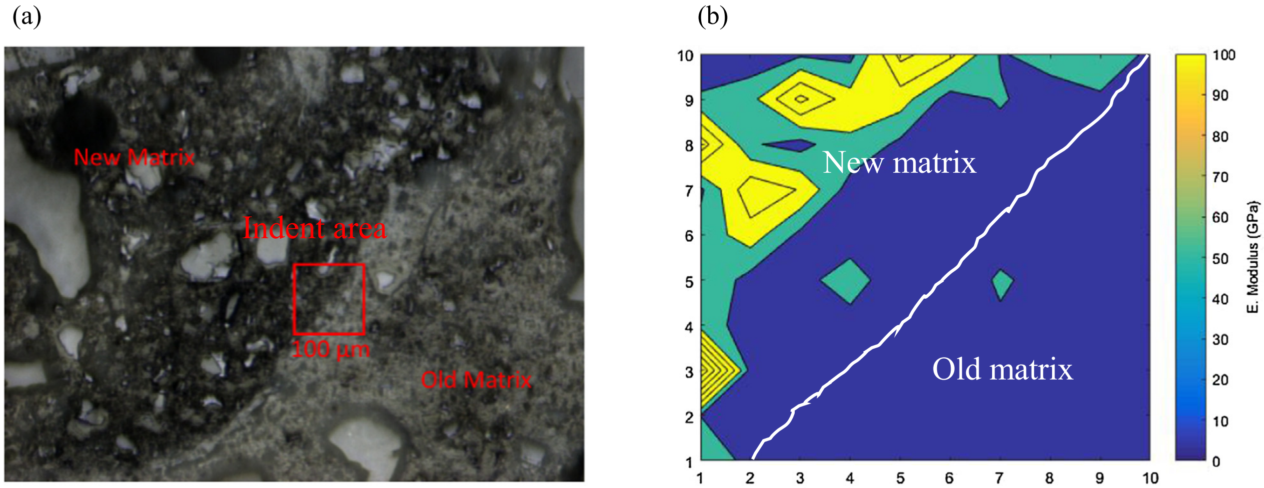

Fig. 16. Nanoindentation of the mortar sample made with the pristine RCA: (a) optical microscope image; (b) contour map of elastic modulus where x and y are coordinates of the indent locations.

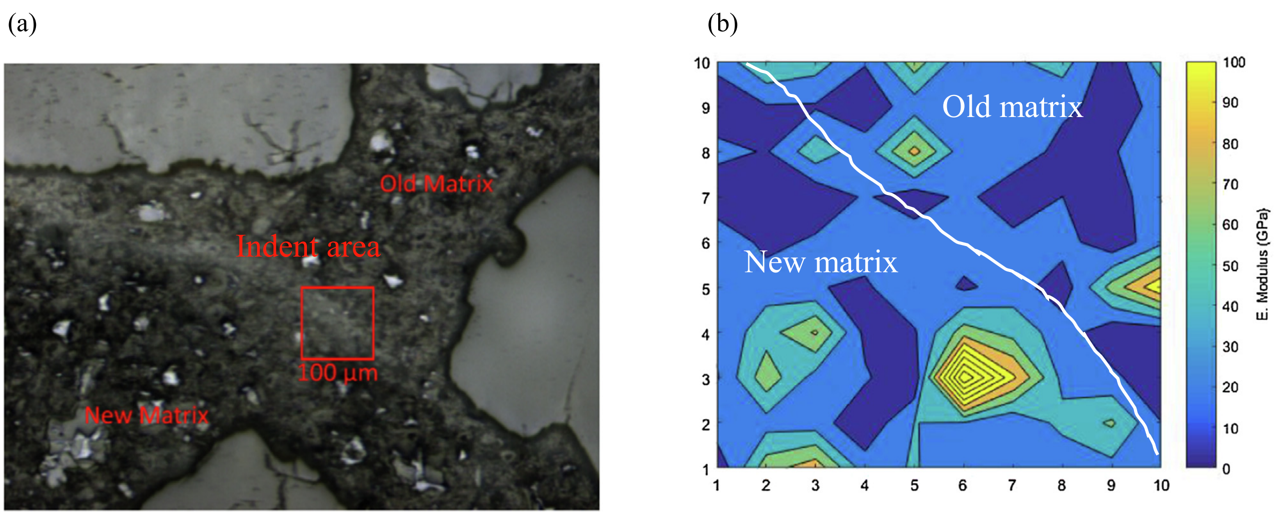

Fig. 17. Nanoindentation of the mortar sample made with RCA treated with 0.5% TA: (a) optical microscope image; (b) contour map of elastic modulus where x and y coordinates represent indent locations.

TA did facilitate the hydration of the cement to produce more hydration products locally. A higher elastic modulus in this zone also suggests that better bonding between the new matrix and the RCA was achieved by the TA treatment, which also contributes to higher strength of the mortar.

4. Conclusions

This study examines the effects of treating fine RCA with a tea-stained inspired naturally occurring compound, tannic acid on the performance of the produced cement mortar. Experimental evidence suggests that TA can react with CH and calcite on the surface of the fine RCA to produce nanoparticles. This reaction not only densified the microstructure of the RCA, but also removed some loose particles from the RCA. Consequently, performance of the produced cement mortar with the treated RCA has been significantly improved. Up to 25% improvement on the compressive strength at 90d and 18% reduction on drying shrinkage at 5d of the cement mortars have been achieved by the proposed method. Two reasons are responsible for this improvement: the lower porosity and better bonding between the RCA and the new cement paste induced by the TA treatment. MIP analysis revealed that TA treatment can reduce the capillary pores with size between 10 nm and 100 nm by half, due to the filling effect of the in-situ produced nanoparticles. Nanoindentation testing results confirm that TA coated on the surface of RCA can facilitate local hydration of cement to produce a denser and stronger bonding area between the RCA and the new cement paste.

This study shows that naturally occurring compounds such as TA can be used to treat RCA. Although natural products such as TA are ubiquitous, they are largely ignored in current concrete research. There are many advantages of using natural products such as TA in concrete. They are abundant, renewable, safe, yet low-cost. The commercial extraction of TA from many plant parts is relatively cheap. In addition, TA can even be made from current industrial waste and byproducts.

CRediT authorship contribution statement

Liang Wang: Methodology, Investigation, Writing - original draft. **Jialai Wang:** Conceptualization, Writing - review & editing, Supervision. **Xin Qian:** Investigation. **Yi Fang:** Investigation. **Peiyuan Chen:** Investigation. **Atolo Tuinukuafe:** Nanoindentation test.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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