XVI International Conference on Durability of Building Materials and Components
DBMC 2023, China
K.F. Li and D.P. Fang (Eds)

Application of Nanomaterials for Enhancing the Performance of Recycled Aggregate Concrete-A Short Review

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Abstract. *Incorporation of recycled concrete aggregates (RA) produced from construction and demolition wastes in concrete contributes to the sustainability from two perspectives: reducing environmental pollution and reserving natural resources by reducing the consumption of natural aggregates (NA). However, comparing to NA, RA has lower mechanical properties and higher water absorptivity due to the old mortar attached on RA surface, resulting in weakened interfacial transition zones (ITZs) in the concrete and the consequent reduced durability of concrete when RA was used to replace NA for producing recycled aggregates concrete (RAC). Therefore, extensive research work had been devoted to the enhancement of RAC performance and nanomaterials have demonstrated great potential in this regard. This paper reviews recent progress on the application of various nanomaterials for improving the microstructure as well as nano/mechanical properties of ITZs in RAC, and special attentions were given to the dispersion strategies for nanomaterials which determines the amount of nanomaterials required to achieve reliable improvement in RAC performance and thus the cost of using nanomaterials in RAC.*

Keywords: *Recycled Concrete Aggregates, ITZs, Durability, Nanomaterials.*

1 Introduction

The application of recycled aggregate concrete (RAC) contributes to the sustainability from two perspectives: reducing environmental pollution by recycling the construction wastes and reserving natural resources by replacing natural aggregates (NA) with recycled aggregates (RA). However, comparing to NA, RA has lower mechanical properties and higher water absorptivity due to the old mortar attached on RA surface, resulting in weakened interfacial transition zones (ITZs) in the concrete and the consequent reduced durability of concrete (Wang et al. 2021). Therefore, extensive efforts had gone into enhancing RAC and nanomaterials/nanoparticles (NPs) have demonstrated great potential in this regard. The NPs available as additives for concrete are classified into two categories: active and inert materials. Inert NPs mostly enhance concrete properties through the nucleation and filler effects (Kong et al. 2018, Ouyang et al. 2017), including graphene nanoplatelets (GNPs), carbon nanofibers (CNFs), carbon nanotubes (CNTs), graphene oxide (GO), nano-TiO_2 (NT), and nano-Fe_2O_3 (NF), whereas active NPs affect cement hydration along with the nucleation and filler effects, including nano-SiO_2, nano-$CaCO_3$, nano-clay, nano-metakaolin and nano-Al_2O_3 (Yoo et al. 2022).

GNPs, CNFs, CNTs and GO are all carbon-based nano-materials. Previous studies (Alatawna et al. 2020, Camacho et al. 2014) comparing the effect of CNTs and GNPs on the fresh properties of cement pastes indicated that the incorporation of CNTs above a certain amount could lead to reduction in the workability due to the entanglement of CNTs, whereas better workability was observed when GNPs were used, attributing to a self-lubrication effect. Different from other NPs, the lateral size of GNPs and GO is likely to have prominent effect on their efficacy in enhancing the performance of RAC. The polymerization degree and mean molecule chin length of C-S-H gels show increasing tendency with the increase of graphene dosage and lateral size. Also, the compressive strength, toughness and three-point bending modulus of UHPC increase with the increase of graphene lateral size and dosage up to 0.5% (by mass of binder), but the GO with lateral size of 10 μm shows the largest enhancement effect on three-point bending strength (Dong et al. 2020).

Chen et al. (2012) confirmed that the nano-TiO_2 acted as a catalyst to accelerate the early age cement hydration and thus improved the early age compressive strength, whereas the 28-day compressive strength was found to decrease with the addition of nano-TiO_2. Ali et al. (2010) concluded that the partial replacement of cement with 1% nano-Fe_2O_3 improved the split tensile and flexural strength of concrete but decreased its setting time. Nano-SiO_2 (NS) is the most widely used nanomaterial to improve concrete performance, especially durability. However, the high activity and nucleation of nano-SiO_2 can result in reduction in setting time and slump, and the shrinkage can increase with the use of nano-SiO_2 (Zhuang et al. 2019). Zheng et al. (2021) reviewed the durability of NS and basalt fiber modified RAC and concluded that although the combination of NS and fiber can effectively improve the interface structure and durability of RAC. The conduction calorimetry results indicated that the addition of nano-$CaCO_3$ had an acceleration effect on the hydration of C_3S as well as on the hydration of OPC (Sato et al. 2010). Nanoclay can be considered as a pozzolanic material because its major components are silica and alumina (Abdalla et al. 2022). The total porosity was found to be reduced by 19% after the incorporation of 1% nanoclay (Fan et al. 2014), and a 66%-67% reduction in the chloride diffusion coefficient was observed after the incorporation of 1% nanoclay in RAC. Nano-metakaolin and nano-Al_2O_3 participate in the secondary hydration reaction to increase the amount of C-S-H gel thus the compactness and the compressive strength (Zhang et al. 2021). The compressive strength of RAC with 5% nano-metakaolin and 30% RA was increased by 17.0% as compared to the counterpart without nano-metakaolin, while the efficacy of nano-metakaolin decreased with increasing the content of RA (Xie et al. 2020).

This paper reviews recent progress on the application of nanomaterials for improving the microstructure as well as nano/mechanical properties of ITZs in RAC, and special attentions were given to the dispersion strategies for nanomaterials which determines the content of nanomaterials required to achieve reliable improvement in RAC performance. It should be clarified that although most of the previous studies reviewed herein are about the application of NPs in cement past or normal concrete without RAC, the conclusions obtained in these valuable studies apply to RAC.

2 Dispersion Method of NPs

The poor dispersion of NPs in aqueous solutions is one of the challenges that hinder the widespread application of NPs in concrete production. The difficulty in dispersing NPs arises from their high surface energy and strong van der Waal forces, and the hydrophobic nature for the case of CNTs and GNPs. NPs are generally dispersed into concrete/mortar by adding NPs suspensions during mixing. For the preparation of NPs suspensions, NPs along with a surfactant are added into water and then the mixture is ultrasonicated or mixed for a specific period of time. Jiang et al. (2021) examined the mechanical properties of GNPs-reinforced concrete prepared with different NPs dispersion techniques, and a wet dispersion method combining ultrasonication and higher shear mixing in a mixture of water and polycarboxylate-based superplasticizer was proposed. In that study, the lateral sizes of GNPs obtained by analyzing the optical microscopy images were used to evaluate the influence of different dispersion parameters, and found that both increasing the duration of high shear mixing and combining shear mixing with 15-minute ultrasonication reduce the mean flake size of GNPs. However, the change in GNPs sizes was not related to the efficacy of GNPs in improving the mechanical properties of concrete.

Douba et al. (2023) recently introduced a novel nano-coating dry dispersion method and compared it with the conventional wet dispersion method. In the dry dispersion process, cement is added during the preparation of NPs suspension in ethanol. Then the cement-NPs-ethanol suspension was dried in a drying oven at 105 ℃ for 24-72 h to remove the ethanol, and finally the cake of cement and nanomaterials was crushed to a powder for use. It was shown that the variation in the strength development due to the change of dispersion method depends the NPs type. Dry dispersion method seemed to be more favorable for nanoclay, while sonication is suggested for silica and calcium carbonate particles.

3 Effect of NPs on RAC Performance

Huseien (2023) reviewed the efficacy of NPs in improving the 28-day compressive strength of concrete, and it was shown that the dosage of NPs is generally lower than 6 wt% by mass of binder, and there is no consistent trend with regards to the variation of compressive strength with raising the dosage of NPs. According to the results from a previous study (Xie et al. 2020), where the effect of nano-silica and nano-Al_2O_3 on the property of RAC was examined, it is possible that NPs were invalid or adverse for RAC, because the addition of NS could result in a reduction in the workability which consequently leads to poor consolidation.

There are two methods for preparing NPs-enhanced RAC: pre-soaking RA in NPs suspensions and the direct addition of NPs suspension as for the normal concrete (section 2). Zeng et al. (2020) verified that pre-soaking RC in commercial colloidal nano-silica suspension for 1 hour can effectively improve the corrosion resistance of RAC and postpone the corrosion-induced cracking, and the improvement was attributed to the optimized ITZ by the penetrated NS as indicated from the microhardness tests results. Wu et al. (2018) used the GO suspension prepared using an ultrasonic homogenizer as the mixing water to improve the mechanical property of RAC incorporating recycled fine aggregates and the electrochemical results revealed that the charge transport between the solid and liquid phases was inhibited because the GO refined the pores ranging from 0.04 μm-0.1μm. Allujami et al. (2022) found that 0.1% muti-walled carbon nanotubes (MWCNT) is effective in increasing the compressive strength and impact resistance of RAC, whereas at higher MWCNT doses the agglomeration tends to result in reduction in the efficacy of MWCNT in improving the property of RAC.

4 Quantification of Dispersion of NPs

The dispersion of NPs is one of the key parameters that determine the efficacy of NPs in improving the property of RAC. However, there seems to be a lack of method to evaluate the dispersion state of NPs in the hardened matrix. For the carbon-based nanomaterials, the identification of NPs with the aid of scanning electron microscope (SEM) and energy dispersive spectroscopy (EDS) mapping is relatively easier to the identification of other NPs like nano-silica and nano-$CaCO_3$ which have the same elements with the cement matrix and are also likely to involve in cement hydration (Stephens et al. 2016, MacLeod et al. 2019). Sargam and Wang (2021) proposed a method for quantifying the dispersion of nano-silica in hardened cement matrix through combining SEM-EDS and image analysis-based methodology. Nano-silica particles in hardened cement pastes were identified according to the Ca/Al (8-12) and the corresponding Si/(Ca-S) (1.0-2.0) atomic ratio. Thereafter, the dispersion of NS particles in the binary images was characterized using the distribution integral (Tyson et al. 2011) and quadrat (Haslam et al. 2013) method. However, the correlation between the dispersion of NPs and the efficacy of NPs in improving the properties of concrete was absent in current studies.

5 Conclusions

The nanomaterials or nanoparticles (NPs) are generally dispersed in concrete fresh mix using suspensions prepared by sonication in water or in a mixture of water and dispersing agent. Pre-soaking recycled aggregates (RA) in NPs suspension is effective in improving the interfacial transition zones (ITZs) of RAC thus the property of RAC. The comparison of dry dispersion of NPs using the nano-coated cement and wet dispersion using NPs suspensions revealed that the influence of dispersion method on the efficacy of NPs depends on the type of NPs and the wet dispersion is more effective between the two methods for most of the NPs.

Despite the extensive research work devoted to examining the efficacy of NPs in improving the property of concrete, no consistent trend regarding to the change of compressive strength with the addition of NPs and the dosage of NPs was observed. This could be attributed to the poor dispersion of NPs. A good dispersion in a suspension does not necessarily indicate a uniform distribution in the hardened matrix and the agglomeration of NPs could result in reduction in compressive strength, especially at high dosage of NPs. However, there seems to be a lack of method for the quantification of the dispersion of NPs, and the correlation between the dispersion state and the efficacy of NPs is absent in current studies. Therefore, the optimization of the dispersion method and the assessment of the dispersion state in the hardened matrix are equally important for the application of NPs.

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