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Investigation of the interphase between recycled aggregates and cementitious binding materials using integrated microstructural-nanomechanical-chemical characterization

Mahdieh Khedmati[a], Yong-Rak Kim[b,*], Joseph A. Turner[c]

[a] Department of Civil Engineering, 362 Whittier Research Center, University of Nebraska-Lincoln, Lincoln, NE, 68583-0856, USA
[b] Department of Civil Engineering, 362N Whittier Research Center, University of Nebraska-Lincoln, Lincoln, NE, 68583-0856, USA
[c] Department of Mechanical and Materials Engineering, W342 Nebraska Hall, University of Nebraska-Lincoln, Lincoln, NE, 68583-0856, USA

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ABSTRACT

The mixtures including crushed recycled aggregates have multiple complex aggregate/paste interphase regions compared to conventional concrete mixtures, which brings significant technical challenges in understanding and characterization of their properties. To gain a better understanding of such complex material organization, this study adopted multiscale experimental methods by using nanoindentation test-analysis, laser scanning microscopy, and energy dispersive spectroscopy. The multiscale methods were applied to two different composites in which the same recycled aggregates were mixed with two different cementitious binders: a fly ash-based geopolymer and conventional Portland cement. The test-analysis results demonstrate that, in cement concrete mixtures with recycled aggregates (CCRA), the pre-existing incomplete interphase within the recycled aggregares was observed, although new paste was relatively well-bonded to the old recycled aggregate paste by having an approximately 20-μm thick interfacial transition zone. In geopolymer concrete mixtures with recycled aggregates (GCRA), both the old and new interphase appeared dense. More interestingly, the pre-existing incomplete interphase within the recycled aggregates was filled in the GCRA, which was not the case observed from the CCRA. Further analysis using energy dispersive spectroscopy suggests that geopolymeric materials can reach the pre-existing incomplete interphase and create hydration-geopolymerization products that combine calcium-silicate-hydrate (C-S-H) and sodium aluminosilicate hydrate (N-A-S-H) gel. The resulting cementitious composite is expected to show enhanced mechanical properties owing to a better interphase region.

1. Introduction

Construction and demolition wastes (C&DW) are the excess or waste materials produced during the construction, renovation, and demolition of structures and buildings. The disposal of concrete structures and pavements contributes a considerable fraction of C&DW in many countries and induces environmental burdens [1]. Owing to the high cost of raw materials and natural resources, several studies are being conducted globally to search for new low-cost materials that exhibit durability and good performance. Recycled aggregate is a reusable mixture of aggregate and old cement paste that is generated by removing, crushing, and processing old concrete structures that are considered to be beyond their useful life. Recently, the use of recycled aggregate to manufacture new concrete has gained considerable attention in an attempt to reduce the use of virgin materials, to increase economic benefits, and to promote environmental friendliness. Many studies have investigated the engineering properties of recycled aggregate and the effect of using recycled aggregate on the properties of fresh and hardened concrete [2–7]. It is reported that at a high water-to-cement (w/c) ratio of 0.55–0.75, the strength of concrete with recycled aggregates is comparable to that of conventional concrete even at 75%–100% replacement. However, by reducing the w/c ratio to about 0.4, only 25% reduction in the strength compared to the reference mix was observed [8,9]. The properties of recycled aggregates acquired from crushed concrete have a significant effect on the mechanical properties of high-strength concrete [10].

The production of Portland cement, which is the primary component of concrete, is responsible for high levels of CO_2 emissions, and the cement industry alone contributes to about 7% of the entire global CO_2 emissions [11]. One of the efforts to reduce greenhouse gas emissions and to produce more eco-friendly concrete is the use and/or development of inorganic alumina-silicate materials (e.g., geopolymer),

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* Corresponding author.

E-mail addresses: mahdieh.khedmati@huskers.unl.edu (M. Khedmati), yong-rak.kim@unl.edu (Y.-R. Kim), jaturner@unl.edu (J.A. Turner).

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synthesized from industrial by-products such as fly ash that is rich in silicon and aluminum [12]. In geopolymerization, the raw material is transformed into a paste by using a high alkaline solution which is usually a combination of sodium or potassium silicate and sodium or potassium hydroxide [13]. Geopolymer binders can deliver approximately 80% less CO_2 emissions compared to ordinary Portland cement (OPC) [14,15]. Fly ash-based geopolymer paste can be used as an alternative to cement paste for producing concrete, since it can significantly decrease the greenhouse gas emissions and consume a large amount of industrial wastes.

Many studies have investigated the properties of fly ash-based geopolymer and shown that geopolymer has advantages of low creep, high compressive strength, strong bonding to the aggregate, low shrinkage, excellent resistance to sulfate attack, high acid resistance, and great fire resistance [16–20]. Nanomechanical and microstructural characterization of the aggregate-paste interphase in geopolymer concrete mixtures made with natural aggregates have shown that the aggregate surface and paste are usually bonded together tightly due to the formation of sodium aluminosilicate hydrate (N-A-S-H) gel, which is the main reaction product of fly ash-based geopolymer [21,22]. It is reported that, in conventional OPC concrete, there is a weak zone between the aggregate and cement paste which is prone to damage and degradation and is called interfacial transition zone (ITZ) [23–27].

As illustrated in Fig. 1, concrete mixtures including crushed recycled aggregates have multiple complex aggregate-paste interphases compared to conventional concrete mixtures, which leads to significant challenges in understanding and characterization of the concrete properties. There are a number of aggregate-paste interphases within a single recycled aggregate, and the recycled aggregate adheres to a new paste. This organization subsequently leads to different interphases: old interphases between the old aggregates and the adhered old cement paste, and new interphases between the old cement paste and the new cement paste [28,29]. The failure behavior of the concrete mixtures with crushed recycled aggregates depends on the relative quality of the old and new interphases [30]. The lower strength is typically owing to the presence of weak ITZs and old adhered paste [29]. Although it is generally reported that the adhered old paste or the old ITZ is the weakest zone [31], other studies have shown that the adhered old paste is not always the main factor controlling the quality of the entire concrete mixtures with recycled aggregates [32], and the relative strength of the old paste and new paste is the main factor that might define the weakest link [33]. Other studies have measured the nanomechanical properties of both old and new ITZs in cement concrete mixtures made with recycled aggregates by conducting nanoindentation and microhardness tests. Based on their results, the old ITZ showed lower indentation modulus than the old paste matrix, and new ITZ also showed lower indentation modulus than the new paste [30,34]. Regarding geopolymer mixtures with recycled aggregates, limited studies have characterized the new and old interphases [36,37]. Liu et al. [35] conducted microstructural characterization of only the new ITZ in geopolymer concrete mixtures with recycled aggregates and found that a weak ITZ does not develop.

2. Research objectives and scope

In this article, the complex interphases between recycled aggregates and cementitious binding materials are investigated in order to improve the understanding of the interaction between two recycled materials, namely fly ash (an industrial by-product) and recycled aggregates. To meet the research objective, recycled aggregates were mixed with two different cementitious materials to produce two concrete mixtures: geopolymer concrete with recycled aggregates (GCRA hereafter) and cement concrete with recycled aggregates (CCRA hereafter). This study used multiscale experiments (spanning the nano-to-micro scales) by integrating microstructure examination using a laser scanning microscope (LSM), nanomechanical characterization using a nanoindentation test-analysis, and spatial mapping of chemical elements using energy dispersive spectroscopy (EDS).

3. Materials and sample preparation

3.1. Materials and mixture ratios

In this study, low-calcium fly ash (Class F) with specific gravity of 2.37 obtained from Boral, Colorado was used as an aluminosilicate source material for synthesizing the geopolymer. The fly ash chemical composition is presented in Table 1. To activate fly ash particles, an alkaline activator solution was prepared by blending sodium hydroxide (NaOH) and sodium silicate (Na_2SiO_3) solutions. Sodium hydroxide pellets with a purity of 98% were dissolved in distilled water to prepare sodium hydroxide solution of 12 molar concentration. The chemical composition of the sodium silicate solution was 9% Na_2O, 28% SiO_2, and 63% water. The activator solution was allowed to equilibrate for a minimum of 24 h at room temperature prior to use. The mass ratio of the NaOH to Na_2SiO_3 solutions was 1.0. As a counterpart of the fly ash-based geopolymer paste, Type I OPC, with chemical compositions as listed in Table 1, was used to produce cement paste. The water absorption capacity and specific gravity of the recycled aggregates were 6% and 2.40, respectively. Recycled aggregates retained on 4.75, 9.5, and 12.5 mm sieves were used in the mixes.

The ratio of alkali solution to fly ash was 0.4 for fabricating GCRA specimens. The fly ash, recycled aggregates, and alkaline solution content were 706 kg/m^3, 1200 kg/m^3, and 282 kg/m^3, respectively. To prepare GCRA, first, the recycled aggregates in saturated surface dry condition and fly ash were blended for 3 min. Afterward, the alkaline solution was added to the blend of recycled aggregates and fly ash. The obtained mixture was mingled for another 5 min and then was cast in cylinder mold. A curing process at 60 °C in an oven was employed for the GCRA specimens for 24 h. Then the demolded specimens kept at a temperature of 23 ± 2 °C for 28 days. The CCRA specimens were

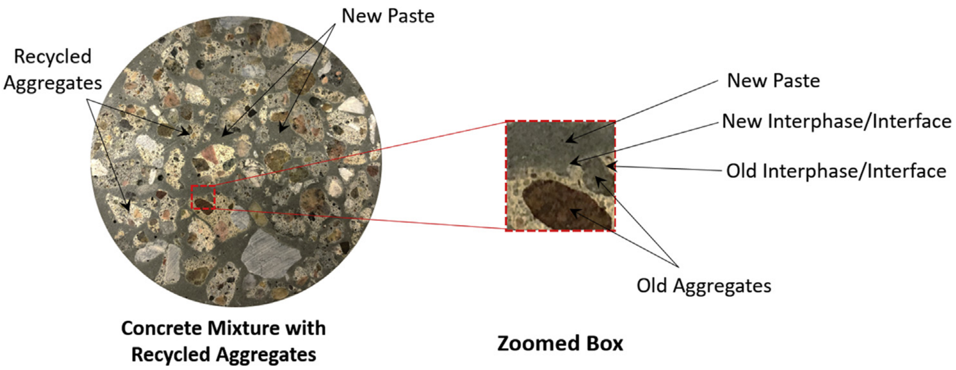

Fig. 1. Schematic illustration of concrete mixture with recycled aggregates.