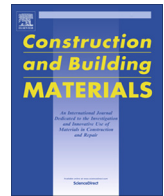




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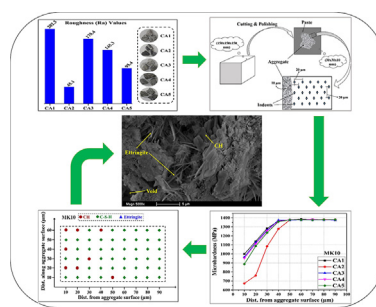
Influence of the surface roughness of crushed natural aggregates on the microhardness of the interfacial transition zone of concrete with mineral admixtures and polymer latex

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HIGHLIGHTS

- Influence of surface roughness of crushed natural coarse aggregate with and without SCMs on ITZ was investigated.
- Aggregate with higher surface roughness indicated higher microhardness values.
- Microstructure enhancement was greatest for metakaolin, followed by silica fume, slag, fly ash, and finally latex.

GRAPHICAL ABSTRACT



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ABSTRACT

Aggregate surface roughness has a significant effect on the interfacial transition zone of concrete. For this study, concrete specimens were prepared from five crushed natural coarse aggregates with varying surface roughnesses obtained from five different sources. The influence of polymer latex and mineral admixtures such as metakaolin, silica fume, slag, and fly ash were also studied. The effect of surface roughness on the interfacial transition zone was characterized by microhardness testing. The microhardness test results were validated via energy-dispersive X-ray spectroscopy (EDS) and scanning electron microscopy (SEM) analysis. The results showed that surface roughness has a prominent effect on microhardness of the transition zone and the effect significantly increases with the addition of cementitious materials. Polymer latex showed a slight influence on the microhardness while the effect of cementitious materials was found to be profoundly increased.

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1. Introduction

The most widely used construction material in the world is concrete, generally a triphasic material at the millimeter scale consisting of aggregate, matrix and the interfacial transition zone (ITZ). The ITZ microstructure is complex; it is distinct from the rest of

the cement paste and is the weakest of the three phases, making it a governing factor in the overall strength of concrete [1]. The origin of ITZ lies in the so-called “wall effect”, due to the uneven packing of dry cement grains against relatively large and flat aggregate surface, resulting in an increased porosity in the vicinity of the aggregate surface [2,3]. The microstructural development of ITZ can be described as follows [1,2,4,5]: (i) During the compaction of freshly poured concrete, a water film forms on the aggregate surface, which account for higher water to cement ratio closer to the aggregate surface than away from it; (ii) secondly, the

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dissolution of calcium sulfate and calcium aluminate compounds in the bulk matrix produce calcium, sulfate, hydroxyl, and aluminate ions, which combine to form relatively large ettringite and calcium hydroxide (CH) crystals in the vicinity of the aggregate surface due to higher water to cement ratio. The accumulation of large CH and ettringite crystals creates a porous microstructure that extends 20–50 μm from the aggregate surface. Thus, the ITZ plays an important role in the strength development of concrete, as the aggregates make up 60–70% of the volume of the whole concrete matrix.

Comprehensive research has concluded that the ITZ plays a vital role in the mechanical as well as durability-related properties of concrete [2,6–13]. The durability of a concrete structure is considerably affected by its transport properties, and the transport-related properties of concrete are directly influenced by the properties of the ITZ [10,13]. In addition, the permeability of the concrete increases as the ITZ structure becomes more porous near the aggregate surface. The porosity of the cement matrix is amplified near the aggregate surface, contributing to increased permeability, chloride migration and water vapor diffusivity [6,12]. Furthermore, concrete structures are more susceptible to damage when subjected to freezing and thawing, and the ITZ exhibits considerably more distortion under freeze-thaw conditions compared to the overall cement matrix [8,11]. Moreover, the microstructure of the ITZ governs the mechanical properties of the cement concrete matrix. The compressive strength of concrete has been shown to improve when the ITZ is modified by the incorporation of nanoparticles [7,9].

Mineral admixtures, also known as supplementary cementitious materials (SCMs) such as silica fume, slag, fly ash, and metakaolin, can improve the durability and mechanical properties of concrete. It is generally understood that SCMs act as a microstructural modifier for the concrete, including the ITZ. Several studies have been conducted to improve the performance of concrete by modifying the microstructure of the ITZ [5,7,14–17]. The results showed a reduction in ITZ thickness when a portion of silica fume and styrene butadiene rubber (SBR) latex was used [17]. The reduction was due to an increase in SiO_2 content in the interface acting as a filler and the presence of a super plasticizer such as SBR latex acting as a deflocculating agent [16]. In addition, the increase in SiO_2 near the aggregate surface creates a corresponding reduction in $\text{Ca}(\text{OH})_2$ content, which increases the microhardness [15]. The addition of mineral admixtures to the cement matrix also improves the pore structure and microstructure of the ITZ. The microstructure of the ITZ and the overall pore structure of the cement matrix exhibited far higher compressive strength when nano silica, silica fume, slag, and metakaolin were used as a cement replacement [7,14]. In summary, SCMs are critically important as microstructural modifiers. Polymers also effectively enhance the performance of concrete due to their deflocculating effect. The fracture energy has been shown to increase due to the presence of polymer latex in the interface zone [5].

Mechanical interlocking between the aggregate and cement paste plays an important role in the local properties of the ITZ and depends on the surface roughness of aggregates. Higher aggregate surface roughness enhances the bond strength between cement paste and aggregate [18,19]. The bond strength increases with increased contact area between the aggregate and the cement paste, which depends upon the aggregate surface roughness [20]. The porosity near the surface of the aggregate also decreased with greater aggregate surface roughness [12]. The fracture energy, stiffness, and ductility of the composite increased with the aggregate surface roughness as well [21].

Based on a review of the current literature, both aggregate surface roughness and the addition of mineral admixtures have a

direct impact on the performance of cementitious composites. However, the previous studies used model aggregates with surface roughness given artificially by various mechanical means, which may not be representative of a natural aggregate surface [12,18,21]. Hence, it is necessary to study the influence of surface roughness using crushed natural coarse aggregates with naturally varying surface roughness. Likewise, the combined effect of mineral admixtures and surface roughness has also not been thoroughly investigated. It is generally accepted that mineral admixtures contain finer particles than cement, which may provide better packing near the aggregate surface. In addition, previous studies have focused solely on the effect of aggregate surface roughness on interfacial bond strength. However, the microhardness test is also widely used to characterize the ITZ in concrete, but is rarely used in the previous studies to measure the influence of surface roughness. In this work, we have studied the effect of surface roughness of crushed natural coarse aggregates on the ITZ in terms of microhardness development. Aggregates from five different sources with a wide range of surface roughness variation were used to prepare the representative specimens for various concrete mixes. The effects of mineral admixtures and polymer incorporation on the microstructure were studied with a Vickers microhardness tester. The results of the microhardness test were further validated by energy-dispersive X-ray spectroscopy (EDS) and scanning electron microscopy (SEM) analysis.

2. Experimental

2.1. Materials

Ordinary Portland cement (OPC) complying with ASTM C150 [22] was used for this study. Mineral admixtures used in this study were metakaolin (MK), silica fume (SF), ground granulated blast furnace slag (slag), and fly ash (FA). The physical properties and chemical composition (provided by the retailer) of the cement and mineral admixtures are shown in Table 1. Natural river sand with a maximum size of 5 mm, absorption of 1.05%, and Fineness modulus (F.M) of 2.7 was used as the fine aggregate. Coarse aggregates (discussed in the text as CA1, CA2, CA3, CA4, and CA5) were obtained from five different sources. Aggregates (angular shaped) CA1, CA2, and CA5 were obtained from Ahn-sung, Hwa-sung, and Yong-in area of Gyeong-gi province; aggregates CA3 and CA4 were obtained from Ok-chun and Gong-ju area of Chungbuk and Chungnam provinces of Republic of Korea, respectively. Table 2 shows the physical properties and mineralogy origin of the coarse aggregates used. A polymer latex with a total solid content of 49.1% and a mean particle size of 252 nm was used for this study.

2.2. Mix proportions

The concrete mix proportions used in this study are given in Table 3. Concrete mix types MK10, SF10, SG10, and FA10 represent the concrete mixes with metakaolin, silica fume, slag, and fly ash at a 10% replacement of cement, respectively. Polymer latex was used at a dosage of 3% (s/s) cement and indicated as Lat3. The acronyms F.A and C.A in Table 3 indicate fine and coarse aggregates, respectively. Same concrete mix proportion was used for all kinds of coarse aggregates i.e. CA1, CA2, CA3, CA4, and CA5. Concrete cubes of $150 \times 150 \times 150 \text{ mm}^3$ and concrete cylinders of $100 \times 200 \text{ mm}^3$ were cast according to BS EN 12390-2:2009 [23] and ASTM C31 [24], respectively. After casting, specimens were placed in laboratory conditions for 24 h. Subsequently, specimens were demolded and cured in lime-rich water for 28 days.

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