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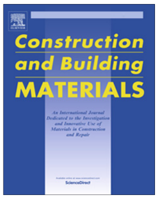

Bonding in cementitious materials with asphalt-coated particles: Part I – The interfacial transition zone

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HIGHLIGHTS

- Backscattered electron microscopy used to study ITZ of mortar with RAP aggregates.
- Larger, more porous ITZ forms from RAP aggregates compared to dolomite.
- Less CH and C-S-H are present in ITZ near the RAP interface.
- Silica fume does not significantly improve ITZ properties in mortar with RAP.

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ABSTRACT

Reclaimed asphalt pavement (RAP), when used as a coarse aggregate, has been shown to reduce bulk concrete strength and modulus. Part I of this study quantifies and compares the interfacial transition zone (ITZ) for mortar with RAP aggregates relative to dolomite aggregates through image analysis of backscattered electron micrographs. The ITZ with RAP aggregates was larger and more porous with less calcium silicate hydrate (C-S-H) and calcium hydroxide (CH) at the asphalt interface compared with dolomite aggregates. The CH morphology was not significantly affected, although the presence of the asphalt layer may be affecting the CH growth. The addition of silica fume reduced the porosity and size of CH particles in the ITZ with RAP, but not sufficiently to be similar to the ITZ of the dolomite mortar. The microstructural changes caused by RAP aggregates, primarily the larger, more porous ITZ, provide strong evidence for the observed reduction in concrete strength and modulus.

1. Introduction

Reclaimed asphalt pavement (RAP) has traditionally been used as recycled aggregate in new asphalt pavements, but has also been identified and studied as an aggregate source for concrete [1–7]. Field demonstration projects have proved the viability of RAP as a virgin aggregate replacement in concrete pavements [8–13]. While there is evidence to support that concrete pavements containing a certain percent replacement of RAP aggregates will have suitable flexural load capacity [2], all previous research studies have indicated that inclusion of RAP aggregates will reduce the concrete strength and modulus [1,2,4–7,14–27]. The poor bonding between the cement paste and asphalt-coated aggregates is believed to be the primary mechanism for reducing the concrete strength and modulus [1,22,26], but no studies have explicitly quantified the microstructural differences of this aggregate-paste interface.

The interfacial transition zone (ITZ) is typically regarded as the region, up to about 50 μm thick, surrounding the aggregate and is characterized by its higher porosity, lower density, lower calcium silicate hydrate (C-S-H) content, lower unhydrated (UH) cement content, larger calcium hydroxide (CH) crystals, and higher ettringite content relative to the bulk paste [28,29]. Quantitative characterization of the ITZ can be performed through image analysis of polished epoxy-impregnated sections using the compositional backscattered electron (BSE) mode in a scanning electron microscope (SEM) [30–32], as has been demonstrated through a number of studies (e.g. [33–44]). Through greyscale thresholding of a compositional BSE micrograph, the ITZ constituents can be quantified, namely porosity, CH, and UH cement. Past research (e.g. [33–44]), has, in general, shown that across the depth of the

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ITZ starting at the aggregate interface, the porosity decreases and the UH cement increases. The CH content has been shown to decrease [37,45] or remain relatively constant [34,35]. The C-S-H and otherwise generally-labeled hydration products have also been found to remain relatively consistent across the depth of the ITZ [38,39], although one study found that the total CH plus C-S-H content decreased across the depth of the ITZ [46].

Bonding at the cement-aggregate interface is both physical and chemical. Indeed the ITZ is often referred to as the "weak link" in the failure of concrete, which is primarily related to three sources [47,48]: (1) the higher porosity in the ITZ, (2) the larger and preferentially-oriented CH crystals, and (3) the potentially weak cement-aggregate bond. To fully characterize how RAP affects the ITZ, all three of these items need to be evaluated. Part I of this study focuses on the physical bond, namely as related to Items (1) and (2) above, while Part II will focus on the chemical aspects of bonding, namely Item (3) above.

Investigations of the ITZ properties in concrete with RAP have not been specifically investigated, although Sachet et al. [49] used SEM with secondary electron imaging to examine fracture surfaces of roller-compacted concrete samples with RAP and noted some potential evidence of poor bonding between the asphalt and the cement matrix. The objective of the present study was to examine the microstructure of cement mortars with RAP aggregates with BSE microscopy in order to quantify how the ITZ composition and morphology are affected by the asphalt coated aggregate particles.

2. Materials and methods

2.1. Cementitious mortar

Mortar was prepared consisting of 40% aggregate and 60% cement paste (Type I Portland cement with water-to-cement ratio of 0.42) by volume. The aggregates were sieved and washed over a #4 (4.75 mm) sieve to remove fines and dust and then air-dried at 50% relative humidity and 23 °C. As aggregate gradation has been shown to influence the ITZ (e.g. [40]), the gradation was controlled to include particles sized 4.75–9.5 mm. The mortar was cast in 5 cm by 10 cm cylindrical molds, cured for 24 h, and then stored in lime-saturated water at 23 °C until the necessary age. In the silica fume mortars, 10% undensified silica fume was used as a partial replacement (by volume) of cement. The Portland cement composition was primarily 63.0% CaO, 20.7% SiO_2, 5.0% Al_2O_3, 3.2% Fe_2O_3, 2.6% MgO, 2.4% SO_3, 0.5% K_2O, 0.3% TiO_2, and 0.1% Na_2O_3 [50].

Three different RAP sources were investigated in addition to a virgin dolomite aggregate. The three RAP aggregate sources shown in Table 1 consisted of dolomite or steel furnace slag (SFS) aggregates. The effects of these specific RAP sources on the concrete bulk properties were studied previously [1,50,51] along with percent of the aggregates coated with asphalt [52], which are also included in Table 1. The original asphalt performance grade (PG) and the aged asphalt PG grade is also shown in Table 1. Both RAP1 and RAP2 were sourced from dense-graded asphalt pavement mixtures, with RAP2 being a similar but less processed version of RAP1. RAP3 was milled from the surface layer of an asphalt pavement, which contained SFS to improve abrasion and friction.

Table 1
RAP aggregate and concrete properties.

Name	RAP1	RAP2	RAP3
Source Aggregate	Dolomite	Dolomite	SFS[a], Dolomite
Asphalt Content	2.10%	3.30%	3.90%
Percent Aggregate Coating	56%	69%	89%
Original Asphalt Grade	PG 70-22	N/A	PG 76-22
Aged RAP Asphalt Grade	PG 88-22	N/A	PG 82-22
28-Day Concrete Strength Properties (MPa)			
0% RAP	46	37	46
20% RAP	37	33	37
35% RAP	33	31	–
50% RAP	28	28	29

[a] Specifically, this is a basic oxygen furnace slag.

2.2. Sample preparation

The sample preparation methodology and technique in this study was developed based on a number of discussions in the literature [53–58]. After 1, 7, and 28 days of curing, thin sections (~1 mm thick) of mortar were cut with a low-speed diamond saw using isopropyl alcohol as a lubricant. The thin slices were vacuum-dried for 24 h prior to epoxy impregnation. As asphalt is very susceptible to solvents, solvent replacement techniques were not applicable for this material, which is why vacuum-drying was employed. Each section measured 15–20 mm by 15–20 mm and was cast into a 32 mm diameter epoxy puck. A low viscosity epoxy (with a measured viscosity of 330 cP) was used to impregnate the samples. The epoxy viscosity was reduced by 50% (330 cP to 160 cP) with the addition of 5% toluene, which has been shown to improve penetration depths in cementitious samples [43].

After 24 h of curing, the epoxy-impregnated samples were polished for SEM investigation. Successively finer grits of silicon carbide (SiC) grinding papers were used (400, 600, 800, 1200, and P4000) in a dry condition followed by diamond paste down to 0.25 μm on a clean glass plate. Additional polishing details for these samples can be referenced elsewhere [59].

The electron microscopy was performed using a JEOL JSM-6060LV SEM. To ensure conduction, the sides of the epoxy were coated with conductive carbon paint and the top surface was sputter-coated with approximately 75 Å of gold palladium. An accelerating voltage of 12 keV was used. All images were taken at a magnification of 500×, resulting in an 8-bit greyscale image measuring 960 by 1280 pixels. Each pixel measured approximately 0.2 by 0.2 μm. The contrast and brightness settings were adjusted such that the image clearly showed the relevant features of pores, hydration products, CH, and UH cement.

2.3. Image analysis

After digitally removing the aggregate from the image, the greyscale image was thresholded to identify pores, CH, UH cement, and hydration products. Greyscale threshold levels were selected on a per-image basis from the greyscale histogram and image processing. In the past studies, thresholding levels were visually selected for each image [60–63], while other studies have specified a set of threshold levels to identify properties for all images [46,64], which implicitly assumes that the brightness and contrast settings on the microscope remain constant. To deter the potential errors from thresholding, the per-image thresholding levels utilized for this study account for potential changes in the brightness and contrast settings on the microscope. The porosity thresholding was performed following the pore overflow technique by Wong et al. [65], which selects the threshold from the inflection point in the cumulative greyscale histogram, an example of which is shown in Fig. 1a. The UH cement was thresholded based on the local minimum in the greyscale histogram after the hydration product peaks [30,60], as seen in Fig. 1b. While CH been reportedly difficult to threshold [37,62,66], in this study a sigma noise averaging filter [44,60] was used to sharpen the peaks in the greyscale histogram. The CH peak was identified at early ages (see Fig. 1b), and an iterative CH thresholding process was employed at later ages, particularly for the mortar with silica fume. The BSE compositional images

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