



Fine aggregate substitution by granular activated carbon can improve physical and mechanical properties of cement mortars



Ismael Justo-Reinoso, Wil V. Srubar III, Alejandro Caicedo-Ramirez, Mark T. Hernandez *

Department of Civil, Environmental, and Architectural Engineering, University of Colorado Boulder, ECOT 441 UCB 428, Boulder, CO 80309-0428, USA

HIGHLIGHTS

- Sand was substituted by hydrated GAC particles of similar size distribution.
- GAC increased compressive/tensile strength when sand was replaced $\leq 2\%$ by mass.
- GAC particle dose influences pore size distributions in a non-linear manner.
- GAC decreased porosity and critical pore diameters when sand was replaced $\leq 1\%$ by mass.

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ABSTRACT

Porosity and strength responses that result from the substitution of fine sand aggregate with similarly sized granular activated carbon (GAC) particles, were studied in cements commonly used in North America. In addition to changes in density and mechanical properties, pore structure responses were analyzed using mercury intrusion porosimetry (MIP). Increases in both compressive and tensile strength resulted from GAC incorporation, where sand replacement was 2% by mass or lower; porosity and critical pore entry diameter also decreased near this range ($<1\%$). Results suggest that bituminous GAC incorporation into cementitious materials may have beneficial effects within specific sizes and mass substitution ranges.

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1. Introduction

1.1. Literature review

The selective sorption properties of activated carbon have been leveraged for water and air treatment for generations, yet carbon particles have been traditionally considered as a potential contaminant of aggregates in cementitious materials. There have been increasing indications that certain types of activated carbons may confer beneficial properties to cement and concrete. As early as 1952, reports emerged suggesting the utility of using activated carbon as a cement additive in industrial applications to counteract contamination of oil well linings from exposure to drilling muds [1]. A retrospective of more recent attempts to incorporate activated carbon into cement formulations reveal diverse engineering

motivations that include the use of both powdered and granular carbon in the following applications: (i) the concrete walls of roadway tunnels and parking garages for the express purpose of absorbing nitrogen oxides (NO_x) from combustion by-products [2,3]; (ii) delivery of viable bacteria to enable “self-healing” concrete from microbial metabolism [4]; (iii) as a substitute for conventional aggregate to reduce concrete density [5]; (iv) as a cement additive for the targeted adsorption of volatile organic compounds (VOCs) in confined spaces [6]; (v) as a means to reuse waste activated carbon in porous concrete [7], and, (vi) as a biocide delivery agent in concrete sewer pipelines to inhibit microbially induced corrosion (MICC) [8]. Unlike aqueous applications, most attention to activated carbon in the concrete materials sector has focused on powdered activated carbon (PAC) incorporation. Although activated carbon has shown some promising potential to expand the performance of cementitious materials beyond conventional applications, there remains a paucity of information on the influence granular activated carbons (GAC) can have on cured cement structure and strength characteristics [4].

* Corresponding author at: 1111 Engineering Drive, ECOT 441 UCB 428, Boulder, CO 80309, USA.

E-mail address: mark.hernandez@colorado.edu (M.T. Hernandez).

Given the demands for broadening the functions of conventional building materials, there has been renewed attention to changing the passive role aggregates play in cementitious materials. Considering the basic design of cement mortars, compressive strength is the mechanical property which is most often specified, since one-dimensional crushing tests are relatively easy, inexpensive, and widely available as compared to some other material performance tests (e.g., adhesion, tension, durability). Additionally, other mortar properties have been related with, if not co-variant upon, conventional compressive strength data (i.e., elastic modulus [9], tensile strength [10]) [11]. Indeed, the mechanical behavior of cementitious material depends on microstructural characteristics, and there is a consensus of literature which suggests that porosity is a reliable measurement, with repeatable ranges associating with select strength characteristics of common cement-based materials [12]. Not only is pore predominance considered critical in cured cement, but pore structure is considered important as well, particularly where maximizing durability is desired [13]. Cement mortars can have highly heterogeneous microstructures, and substituting any part of the fine aggregate fraction with either PAC or GAC, may change enough microscale material interactions that strength can be significantly affected. Under conventional specifications, fine aggregates are often viewed as having little influence on the strength of common cement mortars; thus, sand has often been relegated to the status of an inert filler in practice. However, conventional perspectives on the passive role fine aggregates play in cementitious materials have been recently reconsidered [10]. In this context, it is important to highlight that the association of aggregates with bituminous carbon (e.g., coals), activated or otherwise, is still considered potentially harmful to concrete formulations, as these types of carbonaceous particles may affect strength, durability, and aesthetics [10]. As judged on a mass basis, ASTM maximum limits on the carbon content of fine aggregate has been established at 0.5%, where the architectural appearance of concrete is a primary consideration, and 1% on all other types of concrete; some other international standards are consistent with such limits [14–16].

Admixtures have a broad diversity of chemical composition and desired functions; they remain an important, necessary component of modern concrete and cement mortar formulations. Any admixtures may behave differently depending on the type of cement, as well as interact with other admixtures. Certainly, this may be the case with the activated carbon particles used herein. Because of the sorptive properties of activated carbon, some interactions with other admixture may be anticipated, while others may not; this anticipation includes, but is not limited to, air-entraining agents and superplasticizers. In this context, there is no information available about the interaction of granular activated carbon particles (GAC) and the tremendous diversity of different cementitious material admixtures that are commonly used in modern formulations.

As defined by the American Concrete Institute (ACI), GAC qualifies as a lightweight aggregate (LWA) where its particle sizes are less than 9.5 mm and bulk densities are less than 1120 kg/m³ [17]. Highly porous aggregates, including GAC, have been reported to change microstructure such that their presence can reduce cement strength [11]. However, microstructure improvements associated with increased hydration of cementitious binders have also been suggested to occur in response to activated carbon additions [13]—presumably through the availability of interstitial water, which can desorb from porous LWAs. Internal curing, as described by ACI 213, refers to the process through which cement hydrates based on the availability of internal water that is not part of the bulk water introduced during initial formulation for workability. This internal water can be made available from lightweight aggregates that absorb and subsequently release water [10,17,18].

Unlike conventional aggregates, relatively high mass fractions of water can be stored in activated carbon particles that are engineered to release moisture during the internal curing process; such water cannot be sorbed or otherwise stored by conventional aggregates. Internal curing enhancements can also serve to minimize the plastic (early) shrinkage that can occur by rapid drying [10], which helps reduce the deleterious propagation of internal microcracking. Consistent with the potential for curing benefits, we report that in certain ranges, substituting conventional fine aggregate (sand) with a porous, lightweight, hydrophilic aggregate (GAC of bituminous origins), has a significant effect on microstructure which can manifest in desirable microstructures and strength characteristics.

1.2. Scope

The purpose of this study was to investigate the influence of using granular activated carbon (GAC) as a fine aggregate substitution on the physical (i.e., porosity) and mechanical properties of standard cement mortars. Sand particles were replaced with similarly sized GAC particles, and the resulting mass proportions were isolated as a process variable. In addition to compressive and tensile strength responses, this study analyzed the influence GAC–sand substitutions can impart on pore (entry) size distribution, as well as the bulk density of standard cement mortar formulations used in the United States (ASTM Type I/II) and Mexico (NMX Type CPO).

2. Materials and methods

2.1. Materials

2.1.1. Granular activated carbon (GAC)

Granular activated carbon (GAC), produced by the Calgon Carbon Corporation (USA) and commercially sold as OL 20 × 50, was used to replace specific masses of fine aggregate in the cement mortar formulations used in this study. The GAC was manufactured from bituminous coal and was activated using superheated steam. This GAC had a minimum iodine number of 1050 mg/g [19] which indicated a specific surface area of approximately 1050 m²/g [20]. The particle size distribution of this GAC was determined by standard sieve analysis, which is summarized in Table 1. Water sorption potential and saturated surface dry (SSD) specific bulk gravity were determined in accordance with ASTM standard C128-15 [21], and presented in Table 2.

2.1.2. Graded standard sand (Ottawa sand)

Ottawa test sand was obtained from the U.S. Silica Company (USA), which is a clean natural sand with high silica content; it contains the following constituents on a percent basis: between 99.0 and 99.9% crystalline silica (quartz), <1.0 aluminum oxide, <0.1 iron oxides, and <0.1 titanium oxides [22]. This sand conforms to ASTM standard C778-15 [23]. The particle size distribution of this sand was also determined from standard sieve analysis, which

Table 1
Mass-based sieve analysis of granular activated carbon (GAC) and Ottawa sand.

Sieve	Sieve size (mm)	GAC Calgon OL20 × 50 Passing by mass (%)	Ottawa Sand Passing by mass (%)
10	2.000	100.00	100.00
20	0.850	99.56	100.00
40	0.425	2.08	69.50
60	0.250	0.61	14.79
100	0.150	0.51	2.86
200	0.075	0.32	0.03

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