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A novel type of controlled low strength material derived from alum sludge and green materials



Lei Wang^a, Fuliou Zou^a, Xiaoliang Fang^a, Daniel C.W. Tsang^{a,*}, Chi Sun Poon^{a,*}, Zhen Leng^a, Kitae Baek^b

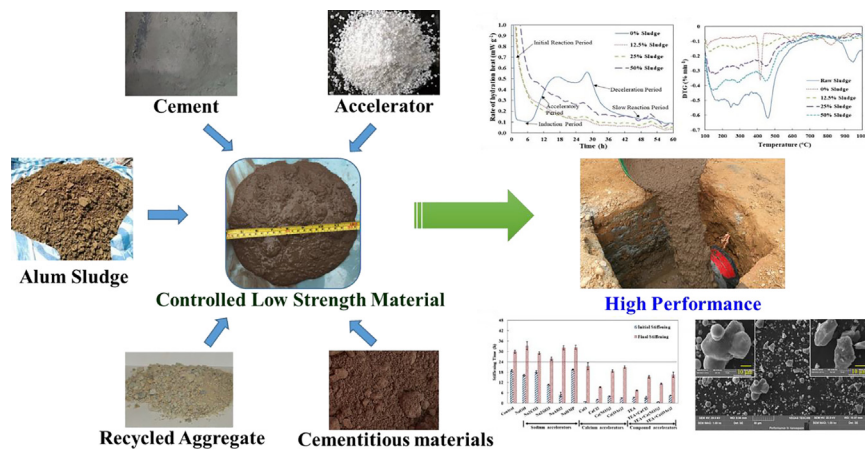
^a Department of Civil and Environmental Engineering, The Hong Kong Polytechnic University, Hung Hom, Kowloon, Hong Kong, China

^b Department of Environmental Engineering and Soil Environment Research Center, Chonbuk National University, 567 Baekje-daero, Deokjin-gu, Jeonju 561-756, Republic of Korea

HIGHLIGHTS

- Alum sludge can be recycled as fine aggregate into eco-friendly CLSM.
- Alum sludge addition increased water demand and hindered CH formation.
- PFA was a promising supplementary cementitious material for producing CLSM.
- Addition of CaCl₂ or TEA effectively shortened overlong stiffening time.

GRAPHICAL ABSTRACT



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ABSTRACT

The management and disposal of alum sludge generated from water treatment works is a worldwide issue. This study proposed an innovative method to utilize alum sludge as a partial replacement of recycled fine aggregate for producing controlled low strength material (CLSM). Various supplementary cementitious materials (SCMs) and accelerators were incorporated to produce CLSM with high flowability (>200 mm), relatively short stiffening time (<24 h), and moderate compressive strength (0.3–1 MPa). Cement chemistry and microstructure characteristics were evaluated by using isothermal calorimeter, quantitative X-ray diffraction, thermogravimetric analysis, and scanning electron microscopy analysis. The addition of alum sludge significantly increased water demand for maintaining sufficient flowability, resulting in long final stiffening time and low compressive strength. The high content of organic matter in alum sludge hampered the Ca(OH)₂ formation and pH increase, thus delaying the induction period of cement hydration. The binder composition played an important role in the stiffening time of CLSM. Pulverized fly ash (PFA) was found to be the most effective SCM for improving flowability and adjusting compressive strength. Calcium-based accelerators or triethanolamine (TEA) significantly shortened the stiffening time to 7.4 h, which facilitated the samples to fulfill all the CLSM requirements. This study presents a new and eco-friendly approach to transform alum sludge into value-added construction materials in a sustainable way.

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* Corresponding authors.

E-mail addresses: dan.tsang@polyu.edu.hk (D.C.W. Tsang), chi-sun.poon@polyu.edu.hk (C.S. Poon).

1. Introduction

The demand of drinking water treatment is increasing along with exponential growth of population as clean water is indispensable for human life. Alum-based coagulants are the most widely used chemical agent for coagulation/flocculation process in surface water treatment works. As a result, alum sludge is massively generated with water purification [1]. The management and disposal of alum sludge has been a global issue. Different from sewage sludge, the low calorific value of alum sludge makes it impossible to recover energy from incineration treatment. Currently, all alum sludge is subjected to energy-intensive dewatering process followed by non-sustainable landfill disposal due to a lack of robust recycling approaches. In Hong Kong, disposal of dewatered alum sludge to landfills is currently 58 tonnes each day and expected to further increase with urban development [2]. Faced with scarce land resources and stringent environmental requirements, recent studies have investigated the utilization of alum sludge in agricultural and horticultural sectors in recent studies [1]. However, there are some limitations of soil applications such as soil acidification, aluminium toxicity, and phosphorus deficiency. Therefore, developing economical and sustainable approaches for alum sludge recycling has significant social and environmental values.

Alum sludge is composed of high contents of Al_2O_3 and SiO_2 with small particle size, which is similar to conventional clay used as construction materials [3]. Previous studies suggested that alum sludge can be incorporated into clay to produce ceramic bricks, but it deteriorated the properties of ceramic products such as reduced flexural strength and increased linear firing shrinkage [4]. A low ratio of sludge replacement should be applied to avoid unacceptable compromise in mechanical strength [5], e.g., over 5% of alum sludge addition deteriorated the mechanical properties and produced low-quality ceramic bricks [6]. The feasibility of recycling alum sludge into light-weight composite was also studied [7], in which every 2% addition of alum sludge increased water-to-cement ratio by 0.01 due to small particle size of alum sludge. It was reported to delay the cement hydration and reduce the compressive strength [8]. Although calcination process improved the microstructure and enhanced the pozzolanic activity of alum sludge [3], the addition of calcined sludge still increased water demand and reduced compressive strength of the material [9]. Another concern is that calcination is associated with substantial energy consumption and carbon footprint. It is imperative to deal with alum sludge in an environmentally-friendly approach.

In this study, we propose to recycle the alum sludge into Controlled Low Strength Material (CLSM), which is a flowable and self-compacting fill material widely used in the backfill of foundation pits, subgrades and pipeline beddings, void filling of subsurface structures, abandoned pipelines and utility vaults, grooves and retaining walls [10]. CLSM offers advantages of easy mixing, easy placing, self-levelling, and fast stiffening. Moreover, it can be easily excavated at any age with conventional digging equipment when future excavation is required [11]. The application of CLSM can reduce labour requirement, expedite engineering works, and lower construction cost. High-quality CLSM should meet strict requirements including high flowability for self-levelling (> 200 mm), relatively fast stiffening for quick loading support (< 24 h), and controlled low strength for easy excavation (0.3–2.1 MPa) [12,13]. In general, high aggregate-to-binder ratio was applied for CLSM production to obtain low strength, and high water-to-cement ratio was adopted to provide sufficient flowability. Large amount of supplementary cementitious materials (SCMs) could be introduced to replace ordinary Portland cement (OPC) to improve flowability and moderate strength [12]. We propose to use alum sludge as fine aggregate in CLSM system to improve

the flowability and adjust the strength. However, high clay content would increase water demand and lead to overlong stiffening time [14]. More importantly, organic matter would complex with Ca^{2+} and form a coating layer, which significantly retarded cement hydration [15,16]. These hurdles make it difficult to produce sludge-derived CLSM.

The binder composition plays a key role in determining the stiffening time of CLSM. Class-F pulverized fly ash (PFA, waste material from coal-fired power plants) is the most commonly used SCM in the production of CLSM. The use of spherical PFA particles could improve flowability and control moderate strength by the effect of friction rolling, micro-aggregate filling, and pozzolanic reaction [17]. But it may consume $\text{Ca}(\text{OH})_2$ (CH) and retard the stiffening time. Other SCMs could also facilitate cement hydration and shorten stiffening time [18,19], for instance, ground granulated blast furnace slag (GGBS, by-product from iron and steel production) contains a high content of calcium and performs as a hydraulic material [20]. The formation of CH during spontaneous hydraulic reaction may relieve the delayed effect by organic matter in alum sludge. Furthermore, incinerated sewage sludge ash (ISSA, waste material from sludge incineration facility) presents mild pozzolanic reaction [21], whereas silica fume (SF, by-product of silicon and ferrosilicon alloy production) consists of microspherical particles that possibly improve flowability and cement hydration [22].

In addition, the incorporation of accelerators is an effective method for expediting cement hydration and shortening stiffening time. Sodium-based admixtures could increase the alkalinity of pore solution, such that high pH could promote the dissolution of calcium silicate and calcium aluminate, resulting in short stiffening time and high early strength [23]. Calcium-based admixtures could provide oversaturated Ca^{2+} ion around tricalcium silicate (C_3S) and tricalcium aluminate (C_3A) to induce cement hydration [24]. Organic accelerators, especially triethanolamine (TEA), could boost the reaction between C_3A and gypsum to form ettringite (Aft) [25]. Combined use of different accelerators may present synergistic or superimposed effect on the reduction of stiffening time.

In order to provide scientific insights for a potential engineering solution to alum sludge utilization, this study aims to: (i) assess the feasibility of using alum sludge as partial replacement of fine aggregate in CLSM system; (ii) evaluate the effectiveness of various SCMs for achieving short stiffening time and suitable compressive strength; (iii) select suitable accelerators for producing sludge-derived CLSM; and (iv) investigate the surface chemistry and microstructure characteristics of sludge-derived CLSM via microscopic and spectroscopic analyses.

2. Materials and methodology

2.1. Materials for CLSM

The alum sludge was obtained from Tai Po Water Treatment Works of the Hong Kong Water Supplies Department. The alum sludge cake contained 55.8% water content, and dried alum sludge contained 21.6% organic matter based on the results of the loss on ignition (LOI) test according to ASTM D7348 (2013) [26]. The major inorganic elements of alum sludge were detected by X-ray fluorescence (XRF) as shown in Table 1. According to the Toxicity Characteristic Leaching Procedure [27] results, the leachability of heavy metals and organic contaminants from alum sludge fulfilled the acceptance criteria for on-site reuse according to the Hong Kong Universal Treatment Standards [28]. The alum sludge mainly consisted of silt and clay with particle size < 0.05 mm. Thus, it could be directly reused to replace a portion of recycled fine aggregate (RA, size < 2.36 mm) to produce CLSM.

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