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Characterization on a cementitious material composed of red mud and coal industry byproducts

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HIGHLIGHTS

- RCC satisfy with the durability testing requirement.
- Amorphous gel-like becomes the dominant structure.
- RCC material meet with EPA TCLP requirement.

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ABSTRACT

This research was to investigate the possibility of incorporating red mud and coal industry byproducts as the raw material for producing cementitious material. Systematic mechanical strength tests were conducted to evaluate the performance of this cementitious material. Results showed that the designed red mud–coal industry byproducts based cementitious material had higher strength in the middle to late curing age (47.5 MPa in 180 days and 48.7 MPa in 360 days) than the OPC control group. The series of durability tests indicated that the cementitious material met with the ASTM requirement. Moreover, the toxicity characteristic leaching tests demonstrated that this cementitious material had good stabilization/solidification ability to bind the heavy metal in the red mud as raw material. Microanalysis revealed that the amorphous gel was the dominant structure of the material at the middle to late curing age, which possibly played a significant role on the heavy metal binding properties through the polymerization during the hydration process of this cementitious material. In essence, this designed red mud–coal industry byproducts based cementitious material not only meet with the physical and mechanical requirements of the ASTM standards, but also meet with the EPA regulation on the environmental heavy metal leaching limitation. This proves the designed material can possibly be used as a clean technology to recycle the red mud from alumina industries and byproduct from coal industries.

1. Introduction

Coal is still considered as the primary energy resource in the world and coal waste from mining system turns out to be one of the greatest amounts of solid waste in coal industries. For the traditional coal waste resources utilization, the coal solid waste is majorly divided into two categories: combustion and non-combustion. For the combustion part, bottom ash and fly ash are considered as major categories of byproduct after the combustion and they have successfully been applied in the cement and construction material in the last two decades [1]. On the other hand, the non-combustion part faces more challenges in the recycling and utilization, because the coal refuse is a low thermal-value byproduct of the coal mining industry and mainly consists of non-combustible rock and some attached carbon materials that cannot be effectively separated [2]. Coal mines in the US generate an estimated 109 million metric tons (120 million short tons) of coal refuse from 600 coal preparation plants in 21 coal-producing states annually [3]. Currently, large volume of coal refuse is still accumulated at the coal mining sites and raise lots of environmental concern.

Red mud is the solid waste residue of the digestion of bauxite ores with caustic soda for alumina (Al_2O_3) production. Approximately 35–40% of the processed bauxite ore goes into the waste as alkaline red mud slurry which consists of 15–40% solids [4,5].

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The chemical and mineralogical composition of the complex industrial waste is widely different, depending on the source of bauxite and the technological process parameters. However, six major oxides in the red mud are CaO, SiO_2, Fe_2O_3, Al_2O_3, TiO_2 and Na_2O [6]. It is estimated that annually 70 million tons of red mud is produced all over the world [7].

Previous studies have been conducted on recycling these two categories of industrial solid waste by activating them into pozzolanic materials. For example, Zhang et al. have successfully recycled red mud and coal refuse into cementitious material by thermal activation at 600 °C [8,9]. Zhang has demonstrated how coal refuse contains good pozzolanic properties after thermal activation [10–12]. The use of coal refuse and red mud in building material has also been reported [13–15]. However, two disadvantages of utilizing coal refuse as cementitious material have been found. The first one is that the cementitious strength of the coal refuse is still low due to the low CaO content in the coal refuse [7]. But the CaO content in red mud is high which is potential mixed with coal refuse together to increase the cementitious performance. Another disadvantage of calcinated coal refuse is in the low flowability. On the other hand, the round sphere particle of coal fly ash has been proven to have good flowability in some construction materials [16–18]. In this paper, a new research has been conducted to investigate a new cementitious material composed of red mud (15%), coal refuse (15%) and fly ash (15%) as its major composition besides the OPC (53%). The scenario of this composition determination is to keep CaO content between 40% and 50% and increase the $(SiO_2 + Al_2O_3)/CaO$ ratio for better durability performance without sacrificing mechanical properties [6,8]. The physical and mechanical properties, durability, microstructure and metal leaching of this cementitious material will be discussed in detail within this paper.

2. Materials and experiments

The raw materials used in this cementitious material contained red mud, coal refuse, coal fly ash, flue gas desulphurization (FGD) gypsum and ordinary Portland cement (OPC). The red mud used in this experiment was the byproducts from an alumina industry in Texas. The coal refuse, fly ash and FGD gypsum were all from West Virginia. US type I/II cement was used in this experiment to provide the proper cohesion and strength values in the early strength. The recipe design of red mud–coal industry byproduct based cementitious material is listed in Table 1. In order to analyze the chemical and mineral composition of the raw materials, X-ray fluorescence (XRF-1700) analyzer and X-ray diffraction (XRD) system (Rigaku Ultimate) were applied. In this designed composition of this material, the coal refuse and red mud were activated at 600 °C for 30 min in a furnace (Lindberg Blue M, Thermo Scientific) before utilizing as raw material in RCC.

2.1. Mechanical properties test

The recipe design of the cementitious material is listed in Table 1. In the strength test, the compressive strength test followed the ASTM C109 (Compressive Strength of Hydraulic Cement Mortars) [19] and the flexural test was according to the ASTM C348 (Flexural Strength of Hydraulic-Cement Mortars) [20]. All mortar samples were mixed according to ASTM C305 (Mechanical Mixing of Hydraulic Cement Pastes and Mortars of Plastic Consistency) [21]. The procedures were performed as described in the standard: all mixing water was placed into the bowl; cement was added to the water and the mixer was started at a low speed (140 ± 5 r/min) for 30 s; aggregate was then added over a 30-s period while mixing at a low speed; mixing was then increased to a medium speed (285 ± 10 r/min) for 30 s. The mixer was stopped for 90 s to scrape down all of the mortar paste followed by mixing for 90 s at medium speed. Finally, the mixer was stopped, and mortar was cast into the mold. Cubic specimens (50 mm × 50 mm × 50 mm) were cast for each mixture for the unconfined compressive strength test. The molds of the flexural test were prisms in size (40 mm × 40 mm × 160 mm). The strength tests were performed at different curing ages (3–360 days). It was necessary to unmold carefully after the first curing day due to the low strength of the specimens which should be cured in a moist cabinet at 95% humidity and 23 °C after unmolding.

Table 1
Recipe design of red mud–coal waste based cementitious material.

(%) By mass	Coal refuse	Red mud	Fly ash	Cement	FGD gypsum
RCC	15	15	15	53	2

2.2. Flowability measurement

In order to measure the flowability of the mortar paste which was just mixed out of the machine, the flowability test was conducted according to ASTM C230 (Flow Table for Use in Tests of Hydraulic Cement) [22] to determine the water content needed for a cement paste sample to obtain a given flow spread of 110 ± 5%. According to these standards, mortar samples from the different mixtures were placed on the same flow table, subjected to 25 repetitions of a standard table drop and the spread diameters of the samples were measured [23].

2.3. Durability test

In this research, freezing–thawing test, chloride permeability test and alkali–silica reaction (ASR) test were completed as durability performance tests according to the relevant ASTM standards.

2.3.1. Freezing–thawing tests

The freezing–thawing tests were performed as described in ASTM C666 [24]. The concrete beams were subjected to 300 freezing–thawing cycles, and the relative dynamic modulus was recorded (at 100, 200, 300 cycles in this test) to represent the ability in resisting freeze–thaw weathering. Relative dynamic modulus of elasticity $Pc = (n1^2/n^2) \times 100$ (where Pc = relative dynamic modulus of elasticity, after c cycles of freezing and thawing; n = fundamental transverse frequency at 0 cycles of freezing and thawing; and $n1$ = fundamental transverse frequency after c cycles of freezing and thawing).

2.3.2. Chloride permeability test

According to ASTM C1202 [25], the chloride permeability test involves monitoring the amount of electrical current (coulombs) passed through a 102-mm diameter by 51-mm thick concrete disc with a potential difference of 60 V DC maintained across the specimen for a period of 6 h. If the number of coulombs passed lies between 2000 and 4000, the chloride permeability of concrete is considered low, and it is considered very low for the 100–1000 range [26–29].

2.3.3. Alkali–silica reaction (ASR) test

The 25 × 25 × 285 mm prismatic mortar bar was used for the ASR test according to ASTM C1260 [30] and ASTM C1567 [31]. After the casting, the prismatic mortar bars were aged in a sodium hydroxide (NaOH) solution at 80 °C continuously for 28 days with intermittent readings of the length change of the bars taken during the course of the test. ASR-related expansions less than 0.10% at 16 days after casting are indicative of innocuous behavior, while those between 0.10% and 0.20% at the same age are indicative of both innocuous and deleterious behavior in field performance; expansion greater than 0.20% at 16 days of age are indicative of potentially deleterious expansion.

2.4. Microanalysis

Scanning electron microscope (Philips XL30 FEG) was used to analyze the microstructure of the specimens at different curing ages. Before the SEM observation, the target samples were first immersed in acetone for 10 days to stop the hydration process followed by drying under vacuum for 7 days.

2.5. Leaching test

The toxicity characteristic leaching procedure (TCLP) was used to determine the mobility the heavy metal according to the EPA-TCLP 1311 procedure. The leaching heavy metals were analyzed using ICP-OES and ICP-AES [32].

3. Results and discussion

3.1. Characterization of raw material and red mud–coal waste based cementitious material (RCC)

Chemical analysis showed that the total carbon in the coal refuse was 3.43%, and the Higher Heating Value (HHV) was only 342 British thermal units per pound (Btu/lb), suggesting that the coal refuse was not suitable for combustion use. Generally, for combustion use, bituminous coals have heating values of 10,500–14,000 Btu/lb on a wet, mineral-matter-free basis; The heating values of sub-bituminous coals range from 8300 to 11,500 Btu/lb on a