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A novel silica alumina-based backfill material composed of coal refuse and fly ash

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ABSTRACT

In this paper, a systematic study was conducted to investigate a novel silica alumina-based backfill material composed of coal refuse and fly ash. The coal refuse and fly ash had different properties under various thermal activation temperatures (20 °C, 150 °C, 350 °C, 550 °C, 750 °C and 950 °C). It is known that a thermal activation temperature ranging from 20 °C to 950 °C significantly increases the flowability and pozzolanic properties of the coal refuse; however, the flowability of fly ash decreases when the activation temperature is higher than 550 °C because of a severe agglomeration phenomenon on its surface. An optimal design for this backfill material was determined to include an activated portion composed of 5% coal refuse at 750 °C and 15% fly ash at 20 °C. This combination yields the best performance with excellent flowability, a high compressive strength and a low bleeding rate. The microanalysis results corresponded well with the performance tests at different activation conditions. In the coal refuse, kaolinite peaks began to decrease because of their transformation into metakaolin at 550 °C. Chlorite peaks disappeared at 750 °C. Muscovite peaks decreased at 750 °C and disappeared at 950 °C. During this process, muscovite $2M_1$ gradually dehydroxylated to muscovite HT. Furthermore, this paper examined the environmental acceptance and economic feasibility of this technology and found that this silica alumina-based backfill material composed of coal refuse and fly ash not only meets EPA requirements but also has several advantages in industry feasibility when compared with hydraulic backfill, rock backfill and paste backfill.

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1. Introduction

The coal industry has been the fastest-growing fuel industry over the last decade because of its role as the primary fuel used for electricity generation [1]. Coal refuse is one of the largest forms of waste from the coal mining industry and is generally defined as a low BTU-value material based on the minimum ash content combined with the maximum heating value [2]. It is estimated that coal mines in the US generate 109 million metric tons (120 million short tons) of coal refuse from 600 coal preparation plants in 21 coal-producing states annually [3]. Presently, two major methods are applied for the utilization of coal refuse: combustion use and non-combustion use. However, the utilization efficiency of coal refuse remains low [2,3].

Backfill refers to any waste material that is placed in voids mined underground for the purpose of either disposal or performing engineering functions [4]. The backfill industry is particularly interested in technologies that reduce the costs associated with backfilling large open stopes [5]. Previous studies on backfill using industrial waste have been conducted in the last decade, including studies on fly ash, blast furnace slag and flue gas desulfurization gypsum (FGD gypsum) [6,7], and reports on developed backfill materials have demonstrated their excellent workability and good mechanical properties [8,9]. Therefore, backfill material acts as a suitable solution to current industrial solid waste challenges [10,11]. However, there have been few studies on backfill material based on coal refuse, although some research has been conducted on the cementitious material composed of thermal activated coal refuse as pozzolanic material. Zhang et al. have successfully recycled red mud and coal refuse into cementitious material by thermal activation at 600 °C [12,13], and Zhang has demonstrated that coal refuse contains good pozzolanic properties after thermal activation [14–16]. In this paper, a systematic study was conducted on a novel, high-performance, silica alumina-based backfill material, which was designed by taking advantage of the pozzolanic property of thermal activated coal refuse and the good flowability of fly ash. Furthermore, a detailed microanalysis is performed to illustrate the mechanism of coal refuse thermal activation and environmental leaching. The results proved the environmental acceptance of this new backfill material.

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Table 1
Chemical analysis of the coal refuse.

LOD 105 °C, % (w/w)	Ash 750 °C, % (w/w)	Volatile matter, % (w/w)	Fixed carbon, % (w/w)	Total carbon, % (w/w)	Organic carbon, % (w/w)	Total sulfate, % (w/w)	Total nitrogen, % (w/w)	HHV BTU/lb
2.21	90.09	7.50	<1	3.43	3.05	0.041	0.10	342

2. Materials and experimental procedure

2.1. Materials

The raw material used in this experiment included coal refuse, fly ash, cement, blast furnace slag and FGD gypsum. The coal refuse was generated by a mining operation situated within the Central Appalachian Coal Basin in southwest Virginia, eastern Kentucky and southern West Virginia. The coal seams located near these operation sites were deposited during the Pennsylvania period and are located in the Pottsville Group from Pocahontas through the lower portions of the Allegheny formation. The fly ash is a typical combustion byproduct from the coal power plant in Tennessee, which was responsible for a catastrophic spill in 2008. According to the American Society for Testing and Materials (ASTM) C618, the fly ash used in this test should be designated as Class F [17]. The FGD gypsum was from Wisconsin, and the cement applied in the experiment was US type I/II, which has sufficient cohesion and strength to form ettringite in the backfill material.

2.2. Chemical, mineral and physical analyses

The chemical analysis was performed using an X-ray fluorescence (XRF-1700) analyzer, and the mineral composition was detected by X-ray diffraction (XRD) with the Rigaku Ultimate. The particle size pattern was measured using sieves and a laser particle distribution analyzer (Mastersizer 2000).

2.3. Thermal activation

Thermal activated oxidation occurred as the raw material (coal refuse or fly ash, respectively) reacted with oxygen in air. Thermal activation experiments were controlled at different temperatures (150 °C, 350 °C, 550 °C, 750 °C and 950 °C) inside a furnace (Lindberg Blue M, Thermo Scientific) for 45 min per experiment.

2.4. Flowability test

According to previous research and an ACI 229 report, ASTM D6103 was used to evaluate the consistency of the controlled low strength material [18]. An open-ended flow cylinder 6 in. (150 mm) in length with a 3 in. (76 mm) internal diameter was used to test the flowability by measuring the spread diameter when the fresh slurry was released from the cylinder onto a flat surface. It is important to immediately measure the largest resulting spread diameter of the backfill material. Then take two measurements of the spread diameter perpendicular to each other. The measurements are to be made along diameters which are perpendicular to one another [19].

2.5. Compressive strength test

For the compressive strength test, 50 mm × 50 mm × 50 mm cube specimens were cast for each mixture as described in previous research [9]. The compression tests were performed on six specimens of various ages (1, 3, 7, 28, 56 and 90 days). This material was carefully demolded for 24 h after it was cast because of its low and late increase in strength. However, before this process, the samples should be placed in sealed plastic bags to retain humidity. Each recipe had 12 replicates in these experiments.

2.6. Bleeding test

A high bleeding rate is often observed in fly ash-based cementitious material because of the spherical shape of the fly ash particle. In this test, as described in ASTM C232, the water bleeding rate was measured according to the amount of water accumulated at different time intervals on the sample surface of an approximately 14 l cylindrical container with an inside diameter of 255 ± 5 mm and a height of 280 ± 5 mm [20].

2.7. Microanalysis

In this experiment, X-ray diffraction (XRD) analysis was conducted using a Rigaku Ultimate, with CuKα radiation, a voltage of 40 kV, a current of 40 mA and a 2θ scan ranging between 5° and 60°. The Nicolet Impact 400 Fourier transform infrared (FTIR) spectroscopy was used to record the spectra with a Nicolet sample processor using KBr-pellets. The microstructures were observed by a Philips XL30 FEG Scanning Electron Microscope (SEM) with energy-dispersive X-ray microanalyses. The thermal gravimetric analysis (TGA) of the coal refuse was conducted between room temperature and 1000 °C at 20 °C/min.

2.8. Toxicity characteristic leaching procedure (TCLP)

The contaminant leaching tests for the raw material and 180 day backfill sample were conducted according to the EPA-TCLP1311-92 procedure. The concentrations of heavy metals were analyzed using Vista-PRO Varian ICP-OES.

3. Results and discussion

3.1. Characterization of raw material

Tables 1 and 2 summarize the chemical composition and physical properties of the raw materials. The total carbon in the coal refuse used in the experiment was 3.43%, and the higher heating value (HHV) was 342 BTU/lb. These results indicated that the coal refuse was not suitable for direct combustion use. The loss on drying (LOD) was 2.21% after being exposed to air at 105 °C for 1 h. The samples were stage ashed to 750 °C and held at that temperature

Table 2
Chemical composition and physical properties of the raw materials.

	Coal refuse	Fly ash	Slag	Gypsum	Cement
SiO_2 (%)	48.77	49.54	36.59	0.76	13.71
Al_2O_3 (%)	14.54	19.11	13.17	0.19	2.66
Fe_2O_3 (%)	11.12	14.79	0.98	0.17	3.27
CaO (%)	4.34	6.23	35.53	42.29	63.39
MgO (%)	1.68	0.42	7.58	0.71	1.23
SO_3 (%)	0.43	0.34	2.03	53.75	2.19
Loss on ignition (%)	9.91	2.43	2.78	1.98	2.6
Specific gravity (t/m^3)	2.69	2.38	2.98	2.91	3.18
Specific surface area (m^2/kg)	564	551	677	579	372