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Prediction of the long-term performance of green liquor dregs as a sealing layer to prevent the formation of acid mine drainage

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ABSTRACT

One of the mining industry's main concerns is the management of waste rock and tailings generated by sulfide ore extraction. Upon exposure of atmospheric oxygen, iron sulfides oxidize generating acidity. Infiltrating water form a metal-rich acidic leachate called acid mine drainage (AMD), that can cause serious environmental problems. Green liquor dregs (GLD) is a material that resists the passage of oxygen and water and could thus be used to seal mine wastes, preventing their oxidation and AMD formation. To enable its use in dry mine waste covers, the long-term efficiency of such GLD sealing layers must be evaluated. In this study, fresh GLD and GLD aged for 3–13 years was collected from two sites and analysed to determine how aging affects its chemical and physical properties. Aged and fresh GLD were very similar with respect to all the properties important in a sealing layer. In particular, there was no evidence of calcite dissolution in aged GLD samples. Aged GLD also exhibited high water saturation (>91%) and chemical stability, both of which are important for effective long-term sealing. The shear strength of GLD deployed in the field increased over time but not sufficiently to ensure the long-term physical integrity of a pure GLD sealing layer. The development of hybrid materials with improved shear strength will therefore be necessary.

1. Introduction

Tailings and waste rock generated by the mining of sulfidic ores must be managed and disposed of in a way that guarantees the protection of the environment as well as human health and safety. A major problem associated with sulfidic mine waste is the formation of acid mine drainage (AMD) a metal-rich acidic leachate formed by the waste's oxidation that causes serious environmental problems [1]. A common technique for preventing its formation is to bury the waste underneath a dry soil cover whose purpose is to prevent oxygen from reaching the waste and thereby retard its oxidation [2]. The dry cover usually consists of a sealing layer with a high degree of water saturation to prevent oxygen influx and low hydraulic conductivity to limit water infiltration, thereby reducing the amount of drainage water reaching the waste. A protective layer is applied above the sealing layer to protect its integrity. Unfortunately, there is a lack of sealing materials that are both inexpensive and capable of efficiently preventing oxygen and water from reaching the stored waste in the long term. Clayey glacial till is often used as the sealing layer [3] when possible but many mines are not located in close proximity to tills with the necessary properties, creating a need for alternative solutions. Recent studies have shown that certain industrial wastes and residues could potentially be useful in this context [4–7].

One material that has shown particular promise in preventing oxygen and water ingress is green liquor dregs (GLD) [8], which is the largest waste fraction generated during the chemical recovery cycle at sulfate pulp mills and is classified as non-hazardous chemical waste by the Swedish EPA [9]. This alkaline inorganic waste contains calcite, brucite, amorphous phases and insoluble solids, and is known to have low hydraulic conductivity ($<10^{-8}$ m/s) and a high water retention capacity [8]. Previous studies have shown it could be a viable alternative to traditional materials for the construction of sealing layers [8,10,11]. However, there is a lack of information on its long-term performance, which is required before it can be considered a general solution.

Four main properties have been determined to affect the long-term performance of a dry cover: hydraulic conductivity, water retention capacity, degree of saturation and physical integrity [12].

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Identifying how these properties changes as the material ages will make it possible to predict a sealing layer's long-term performance at preventing water and oxygen ingress.

In this study, GLD aged for 3–13 years originating from two paper mills was collected from two landfill sites and compared to fresh GLD of the same origin. The aim was to assess how the four key cover performance properties listed above changed as the GLD aged and to thus predict its long-term performance as a barrier to oxygen and water in dry cover applications for sulfidic mine waste.

2. Materials and methods

2.1. Material

Fresh GLD was kindly provided by the Smurfit Kappa (SK) and Iggesund sulfate pulp and paper mills, both of which are located in northern Sweden.

Aged GLD originating from Iggesund was collected at the Iggesund paper mill landfill. GLD has been stored at this site since 1998 in an organized pattern, making it possible to collect GLD of 3, 8 and 13 years of age. Aged GLD (6 years old) originating from SK was sampled at the Rönnskär landfill at Rönnskärsverken, Sweden, where it had been placed as a sealing layer on top of oxidized tailings (applied in 2006). The site had also been used for disposal of snow from large parts of the surrounding industrial area.

2.2. Methods

2.2.1. Sampling

GLD from SK was sampled at a test plot at the Rönnskär landfill using a plastic shovel. In addition, intact soil cores were collected by slowly pushing a 15 cm long cylinder with an internal diameter of 7 cm horizontally into the sealing layer until it was completely filled with GLD. The collected samples were stored at 8 °C until analysis.

GLD from the Iggesund site was collected by drilling using a mobile drill rig (Georig 607, Geotech Ltd, Askim, Sweden). Samples were taken with an auger to enable sampling at different depths and the acquisition of material of different ages. The samples were then placed in diffusion bags. Pictures were taken of frozen GLD collected from the upper part of the landfill.

2.2.2. Physical properties

Water retention capacity (WRC) was measured on aged and fresh GLD in duplicate. The samples were packed into cylinders and saturated from below. The cylinders were then placed on a ceramic plate and pressurized from below using a pressure plate apparatus (Soilmoisture Corp., USA). The volume of the loose samples was calculated from their bulk density, which was determined from the weight of the dried samples (105 °C for 24 h) divided by the cylinder volume. The total porosity of the GLD samples was calculated according to the following equation: Total porosity = (Particle density – Bulk density)/Particle density. The bulk density of the GLD sealing layer at the Rönnskär landfill was measured in the field by means of a Balloon Densometer Test ($n = 1$). A multivolume helium pycnometer (Pycnometer 1305, Micromeritics, Nercross, GA, USA) was used to determine the compact density of fresh and aged GLD originating from Iggesund ($n = 2$) and aged GLD originating from SK ($n = 3$). The compact density was then used to calculate the GLD's porosity (n), pore number (e) and degree of saturation (S_r).

Ground penetrating radar (GPR) is a geophysical technique for imaging the shallow subsurface by transmitting high-frequency electromagnetic waves into the ground [13,14]. GPR surveys were carried out across the Rönnskär landfill to characterize the integrity of the sealing layer. A RAMAC GPR system (Malå Geoscience, Sweden) was used with 800 MHz and 500 Mhz monostatic shielded antennas (Malå Geoscience, Sweden). GPR profiles were obtained by manually towing the antenna along measured survey lines across the landfill. A "hip chain" was used to trigger each measurement and keep track of distance along the profiles. The sampling frequency was 2500 MHz and a trace interval of 2 cm was used. The number of stacking was set to 8. After acquisition, the profiles were subjected to post-survey processing including time zero adjustment, subtraction of DC-shift and dewow, gain function, bandpass filter and background removal using the Reflex2Dquick software package (Sandmeier scientific software).

Cone Penetration Test (CPT) measurements were performed at both landfills according to a standard procedure [15] to determine each tested material's undrained compressive strength (τ_{fu}) and angle of internal friction (φ).

Cylindrical specimens (5 cm × 10 cm) of fresh GLD from Iggesund ($n = 2$) and aged GLD ($n = 3$) collected at the Rönnskär site were tested under uniaxial compression with an air-hydraulic apparatus. Compression tests were carried out at a deformation rate of 1.5 mm/s.

The dry matter content of fresh and aged GLD from Iggesund and SK was determined ($n = 3$) according to the standard method [16]. Paste pH was analyzed on fresh and aged samples ($n = 3$) with a pH meter (Metrohm Ltd, 704 pH Meter, Herisau, Switzerland) as described previously [17].

Hydraulic conductivity tests on fresh and aged GLD ($n = 3$) from SK were carried out with the Constant Rate of Strain (CRS) method according to a standard procedure [18]. Cylinders measuring 15 cm × 7 cm in diameter were filled with GLD. The samples were then placed under a 30 kPa load for 28 days to mimic the conditions encountered in a 1.5 m protective cover. The cylinders were placed in an oedometer and subjected to a progressively increasing induced stress. Drainage was only allowed from the top. The hydraulic conductivity was calculated based on the deformation and the pore pressure from the lower surface. Hydraulic conductivity tests on Iggesund GLD of 0 (fresh), 3, 8 and 13 years of age were performed in duplicate by the Constant Head Permeability (CHP) method according to a Swedish standard [19] and their results were interpreted using Darcy's equation. Water was pressed through the column from below and collected in sampling bottles using a constant water head. The amount of permeated water was monitored continuously. The samples' density was then calculated, enabling their water content to be determined after drying at 105 °C for 24 h.

2.2.3. Chemical properties

Batch leaching tests on fresh GLD from SK and Iggesund were performed in duplicate according to a modification of the Swedish Standard SS-EN 12457–4 [20]. Samples containing 20 g of GLD on a dry matter basis were placed in 250 ml centrifuge bottles (Beckman coulter) and 200 ml of Milli-Q water with pH of 6.3 (matching that of the local snow) was added to achieve a liquid/solid (L/S) ratio of 10. The mixtures were then shaken with an end-over-end shaker for 24 h and centrifuged at 4000 rpm for 10 min, after which 20% of the supernatants were removed and filtered through a 0.22 μm filter. The leachates' pH, electrical conductivity (EC) and redox potential (E_h) were analyzed immediately using a Voltkraft PH-100ATC pH meter, a WTW Multi 350i multimeter (type Level 1 with WTW 323 electrode, Weilheim, Germany), and a pH/ion meter (Radiometer, Copenhagen, Denmark) with an Ag/AgCl electrode, respectively. E_h values were obtained by adding 207 mV to the readings obtained with the pH/ion meter. Chemical analyses were conducted by an accredited laboratory (ALS Scandinavia AB, Luleå, Sweden). After the leachate's removal, the bottles were topped up with additional Milli-Q water to restore