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Novel ecosustainable peat and oil shale ash-based 3D-printable composite material

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ABSTRACT

In this study, a novel peat composite construction material suitable for three-dimensional printing of entire house boxes (walls, floors, ceilings, etc.) was developed. Peat was used as filler to achieve excellent thermal properties. Using peat for this purpose has not been mentioned in the earlier literature because it retarding properties to the cementation process. As a binder, combustion fly ash was used. The retarding properties of peat humic substances on hardening of pozzolanic mixtures were successfully overcome by using silica fume as an additive. The same additive also greatly improved the long setting time of oil shale ash. The mechanism of hardening of the composite material was hypothesized as the following: first, upon contact with water, the pH of pore water exceeded 13 due to the high content of oxides of alkali metals in the ash, which promoted the oxidation of humates into oxalate. Then, alkali metal, aluminum and silicate ions (the latter mostly originating from silica fume) with alumina formed insoluble feldspars leading to a decrease in pH. At pH values lower than 12.4, Ca(OH)₂ became soluble and Ca²⁺ ions migrated to the liquid phase, allowing pozzolanic reactions to occur and calcium oxalate to precipitate. Finally, the residual Ca(OH)₂ reacted with atmospheric CO₂ to form CaCO₃. Test samples were 3D-printed using experimental apparatus, the printing head involved a nozzle delivering wet peat-ash mixture using compressed air. The material was characterized by measuring its thermal resistance (<0.08 W/(m K)), compressive and bending strengths (1.2 MPa and 0.36 N/mm², respectively). Using peat composites in the construction industry allows reducing CO₂ emissions from partially mined peatlands (currently the one of the largest contributors to the total CO₂ emission from Estonia) and increasing the reuse of oil shale ash while providing a cheap and affordable raw material.

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1. Introduction

Three-dimensional (3D) printing is an advanced manufacturing process that produces a target object automatically from a 3D computer-aided design model without any tooling, dies and fixtures. The target object, which can have complex shaped geometry, is manufactured in a layer-by-layer manner by depositing material exactly where it is defined in the digital model [1]. Although the concept of 3D printing has been hovering around for some time now, only recently has the actual use of 3D printing and its adaption into the construction industry started to become a reality. 3D printing is expected to revolutionize the construction industry by enabling the tackling of the main challenges the construction sector is currently facing, including stagnant efficiency (manufacturing industry vs construction industry from 1995 to 2011 (16 years): 1.7 times less added value) [2] and constantly increasing costs. In addition to faster and more accurate construction process,

the advantages of using 3D printing in the construction industry include reduced labor costs, waste generation, health and safety risks, and improved environmental friendliness, providing opportunities for more sustainable and affordable housing. Using 3D printing, no construction formwork is required [3].

The main objective of this study was to develop an environmentally friendly peat and fly ash based printable construction material. Peat has excellent antibacterial and thermal properties [4] (the thermal conductivity of pure peat is between 0.037 and 0.08 W/(m × K)) [5] and is widely available in many regions in the world [6].

The material can be used to form large blocks or entire construction elements (walls, ceilings, etc.). These elements require minimal after processing, the entire house box can be printed in hours or days including walls, floors, ceilings, roof, etc. The other key element of the technology is a mobile 3D printer allowing printing houses using the described material in situ on the building site [3].

Currently, all the 3D-printing technologies are based on concrete/fiber or clay/fiber compositions. Peat (despite the excellent properties) was not used because of retarding properties avoiding its use in

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concrete composites. Likewise, oil shale ashes are used as cement improvers, not the main composite-forming component due to their extremely long setting time.

Problematic issues, the retarding properties of humic substances and long setting time of oil shale ash were solved successfully in the present study.

2. State-of-the-art

2.1. Organic fibers in construction materials

Concrete–organic materials (wood wool, straw, etc. combined with inorganic binders) panels have been widely used for thermoinsulation for many years. The first mineral-bonded wood composite panels utilized a magnesite binder and were developed in Austria in the early 1900s [7].

Peat was used as construction materials for thousands of years [8]. Viking houses were built of bricks cut directly from peatland. Similar constructions were developed later [9] using natural peat blocks. This method is not suitable for industrial construction because of its high labor costs.

Peat was also used as thermoinsulation for many centuries, typically mixed with 5–10% quicklime and used for filling wall and ceiling constructions. Lime increased the stability and hardness of the mixture. Peat filling in walls built using this method hundreds of years ago remained nondegrading [10].

Using peat for manufacturing of concrete–organic materials has not been mentioned in the earlier literature because of its unwanted retarding properties to the cementation process,

2.2. Influence of peat humic acids on cementation reactions

The humic acid generated from the organic remains after long-time decomposition during peat formation is a type of complex organic acid. The fundamental structures are aromatic and cycloparaffinic rings, which have multiple adsorption, complexation and chelation functional groups (hydroxylic, carboxylic, etc.). The humic acid has an intense chemical affinity for group II metal ions (Ca^{2+} , Mg^{2+} , etc.) and reacts with hydrated lime accompanying the formation of insoluble calcium and magnesium humates [11], which can adsorb on the surface of cementation centers, interfering with the hydration of silicates and pozzolanic reactions, i.e., reactions taking place in mixtures of water, $\text{Ca}(\text{OH})_2$ and acidic or amphoteric oxides (Si, Al, Fe) [12]. Moreover, lower pH values caused by high content of humic acids may hinder the dissolution of clay minerals. Nevertheless, the effects of humic acid on the strength development are not completely understood [13].

Similar issues are described in studies of stabilizing peat-containing grounds with cement [14]. There exist numerous factors retarding the cement hydration process in mixtures containing peat. They include:

- Dissolved and colloidal organics, particularly humic and fulvic acids, can physico-chemically retard or prevent the hydration of pozzolanic binders such as those present in cement or combustion ash in binder-peat mixtures. Humic acids react with calcium ions released from calcium hydroxide or other pozzolanic agents to form insoluble calcium humates, thus retarding mixture crystallization and impeding the increase in peat composite strength. Fulvic acids associate more selectively with aluminum-bearing mineral particles [13], leading to the destruction or decomposition of the layered crystal lattice and resulting in the production of an adsorbed layer of fulvates that impedes the further hydration of pozzolanic substances. In addition, fulvic acid may further prevent the formation of the composite structure by decomposing calcium aluminate hydrate, calcium sulfate–aluminate hydrate and calcium ferrite–aluminate hydrate crystals.
- Dissociation of organic acids present in peat causes the mixture pH to drop, decreasing the dissolution rate of acidic components and

gel formation resulting in a slower strength gain in peat–binder composite. A pH value lower than 9 of the pore solution is too low for the formation of secondary cementation products, further limiting or retarding the hardening of the mixture [15].

- The high organic to mineral ratio in peat implies a high water retention capacity and high moisture content, which, in turn, facilitates the adsorption of organic matter on the surfaces of solid particles. This gives a high water-to-binder ratio, which, by itself [12] [15], limits the increment in strength of the mixture and results in lower strength.

Consequently, more binder must be added for achieving of the desired strength. Humic substances in concrete mixture tend to exhibit a threshold effect whereby the quantity of binder used must exceed a certain level before significant hardening is obtained.

3. Materials

3.1. Oil shale ash

3.1.1. Kukersite industrial use

About 15 Mt of kukersite oil shale (which mainly consist of limestone, CaCO_3), is mined in Estonia annually. Most of this (ca. 80%) is used locally in power plants, 20% is used for oil production [17]. Oil shale is relatively low in energy production (calorific value 8.3–9.0 GJ/t), high in mineral content (Table 1 and Table 2 [18,19]) and CO_2 and SO_2 emission (along with the organic matrix, the pyrite fraction is ignited during the combustion process, releasing SO_2).

As shown in Table 2, the inorganic matrix of the oil shale consists mainly of calcite (limestone) and dolomite. During the combustion process of oil shale, partial or near full thermal decomposition of the carbonaceous fraction of inorganic matrix takes place, depending on the combustion temperature and the average size of oil shale pieces. Thermal decomposition releases CO_2 , in addition to the emission from combustion of the organic fraction. There are two methods for firing oil shale used in Narva power plants: direct combustion of pulverized oil shale (PF) and circulated fluidized bed (CFB), which uses oil shale in larger pieces. A low combustion temperature reduces decomposition of the CaCO_3 present in oil shale, thereby reducing CO_2 emissions and improving efficiency in comparison to PF units [20]. However, the CaO content in ash is reduced - an extent of carbonate decomposition (ECD) is 60–75%, compared with approximately 98% in case of PF, where the combustion temperature is 1400–1450 °C. CFB ashes are therefore not suitable for construction materials production because of their very low hardening properties due to low CaO content formed from carbonates' decomposition process. Furthermore, in CFB boilers, metal oxides react with sulfur oxides and the final oxide content of ashes is lower than with PF boilers.

Typical composition of Estonian kukersite oil shale is as shown in Table 1 and Table 2 [18,19]:

3.1.2. Oil shale ash chemical composition

Oil shale ash was obtained from Eesti Energia Auvere Power Plant [16].

The total production of oil shale ash [21] is ca. 5–7 Mt annually, about half of them are CFB ashes.

About 5–6% of CO_2 emitted can be bound back by ashes produced but this process is extremely low in ash fields [22]. Using combustion ash as raw material allows significantly reduce the overall CO_2 pollution

Table 1
Main components of Estonian kukersite oil shale.

Inorganic matrix	67%
Organic matrix (kerogen)	30%
Bitumen	3%

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