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Pelletized Clay Mixtures with Enhanced Thermal Conductivity Intended for the Isolation of High-Level Nuclear Waste.

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INTRODUCTION

Pelletized clay mixtures are being considered as potential materials for the construction of engineered barrier systems (EBS) and seals in the context of deep geological disposals envisaged for the isolation of high-level nuclear waste (HLW) and spent nuclear fuel (SNF). Such type of facility contemplates placing the HLW/SNF encapsulated in a metallic container in horizontal drifts or vertical boreholes excavated in deep and good-quality rocks (i.e. the natural barrier). The empty space between the rock and the canister will be filled with the manmade EBS. One key function of the EBS is to dissipate the heat emitted by the HLW/SNF. An advantage of high-density clay-pellets mixtures is that its emplacement is relatively easy because the clay pellets are directly projected into the openings without the need for compacting them, enabling filling the technical voids typically found in this type of system (e.g., at the rock-barrier interface), or achieving EBS target dry densities (ρ_d) without the need of in-situ compaction. However, clay pellet mixtures produce large macropores (i.e., the inter-aggregate voids between the clay pellets [1]) that strongly affect the thermal conductivity of the buffer system, particularly during the first operational stages. When the EBS is unsaturated. Note that the thermal conductivity of the water and air are around .6 $W.m^{-1}. K^{-1}$ and .025 $W.m^{-1}. K^{-1}$[2], respectively at room temperature. One possible solution to enhance the heat dissipation around the canister containing HLW/SNF is to mix highly heat conductive material (e.g., graphite) with clay pellets.

The design of repositories for HLW/SNF has typically contemplated a maximum temperature of 100 °C [e.g., 3]. Therefore, most of the previous studies investigating the thermal conductivity of EBS have involved temperatures up to 100 °C [4]. In recent years, there has been an interest in optimizing the disposal of HLW/SNF and higher temperatures are being considered, e, g, 140 °C, and higher, up to 200 °C [5]. Furthermore, previous research has mainly focused on the thermal properties of compacted clays This work investigates the thermal conductivity of both a clay-pellets mixture and a thermally enhanced pelletized clay mixture with 10% of graphite (by % of dry mass) at different temperatures up to 140 °C.

MATERIALS

MX-80 bentonite was adopted in this research to prepare high-density pellets (i.e., $\rho_d \sim 2.1$ Mg/m^3). The main chemical compound of MX-80 is silica (SiO_2), with thermal conductivity of ~ 1.1 $W.m^{-1}. K^{1}$.
The clay pellets were prepared by statically compacting MX-80 clay powder with an initial water content of 5.5% in a cylindrical stainless-steel mold (50 mm diameter and 100 mm height) until achieving the target dry density. Then, the compacted bentonite specimen was extruded from the mold and crushed into small and irregular multi-size pellets using a crusher. The industrial graphite was provided from MWI incorporation. The specific gravity of graphite is 1.92 g/cm^3 and its thermal conductivity is around 128 $W.m^{-1}. K^{-1}$

METHODS

The samples were prepared at an initial $\rho_d \sim 1.50$ g/cm^3, which is in-between the minimum (~1.45 g/cm^3) and maximum (~1.70 g/cm^3) emplacement dry densities that are being considered for this type of project. Alfred's backing method [6]was followed to achieve the target ρ_d (~1.5 g/cm^3) for the multi-sized pellets and to avoid segregation in the sample. Alfred's equation:

$$CPFT(\%) = \frac{D^q - D_s^q}{D_l^q - D_s^q} * 100\% \qquad (1)$$

where CPFT is the cumulative percentage finer than particles with diameter D; D_L is the maximum particle diameter (i.e., 10mm); D_s is the minimum particle size (.03mm), and q is the distribution coefficient (i.e., 0.29). Fig.1 shows the grain size distribution curve (GSDC) of the selected material.

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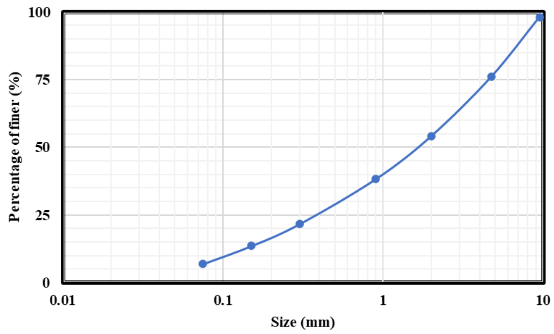

Fig. 1. Grain size distribution curve of the selected material

Three identical samples were prepared in cylindrical aluminum cells (7 cm diameter and 15 cm height) at the target ρ_d 1.50 g/cm^3 and an initial degree of saturation ~ 18%. Two samples were made of pure clay pellets and the third one is a clay pellet: graphite mixture (9:1), by dry mass. The three cells were placed inside an oil bath (Fig. 2) where the temperature was controlled and increased by stages, namely: ~ 24 °C (i.e., room temperature), 40 °C, 60 °C, 80 °C, 100°C, 110 °C, 120 °C, 130 °C, and 140 °C. The cells were sealed using O-rings and high-temperature silicon to prevent any intrusion of the oil into the cells.

Two cells (i.e., one of the pure clay-pellets sample and the thermally enhanced sample) were equipped with a TR-3 needle to measure the thermal conductivity during heating using a 'Tempos thermal properties analyzer' using the transient line heat source method. Three readings were taken by the needle after the samples reach equilibrium for each temperature. The third sample (i.e., made up of pure clay pellets) was instrumented with a thermocouple to check the evolution of temperatures during the experiments.

Fig.2. Adopted setup to measure thermal conductivity at different temperatures.

RESULTS AND DISCUSSION

Fig.3. shows the evolution of the thermal conductivity of the pure clay-pellets mixture and clay-pellets:graphite mixture samples at different temperatures. In both samples, from room temperature up to 120 °C, , the thermal conductivity tends to increase almost linearly with the increment of temperature. This can be explained because the thermal conductivity of each phase (i.e., solid, liquid, and gas) tends to increase with the increased temperature [2] leading to an overall increase in the entire sample. However, a clear decrease in the thermal conductivity is observed with the increase of temperatures beyond 120 °C. This response can be explained by considering the possible evaporation of pore water at elevated temperatures. It is also evident that graphite enhances the thermal conductivity of clay-pellets mixtures by around 60% .

CONCLUSIONS

This work focused on the thermal conductivity of pelletized clay mixtures intended for the construction of EBS. Particularly, the behavior at elevated temperatures has been investigated, expanding the almost inexistent database in this area. The research also contemplated the enhancement of the thermal properties of the clay mixtures by adding highly thermally conductive materials. Specifically, the thermal conductivity of a clay-pellets: graphite (9:1) mixture by dry mass was studied. It was observed that the thermal conductivity of both samples increased almost linearly with the increase of temperature until 120°C, beyond this point a substantial drop of the thermal conductivity with the increase of temperature was registered. It can also be concluded that the addition of graphite effectively contributed to the increment of the thermal conductivity of the pelletized clay mixtures.

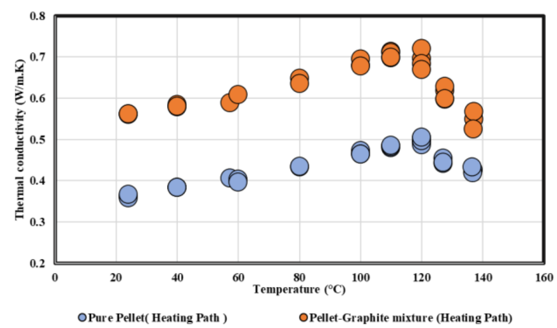

Fig.3 Thermal conductivity at different temperatures

ENDNOTES

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