#### Customized Bentonite Pellets – Extrusion Manufacturing and Gap Filling & Thermal Performance Properties – 14262

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# ABSTRACT

The bentonite pellet related interest in Finland has been focused to provide knowledge about their utilization in deposition of HLW within the KBS-3V concept. Bentonite pellets are intended for use in gaps between compacted bentonite and the rock walls in both buffer deposition holes and tunnel backfilling. The work was concentrated on laboratory experimental tests to optimize the pellet filling by customizing the raw materials and pellet manufacturing processes. Also the measurements of thermal properties of the pellets were emphasized. The work done using extrusion pellets showed that it was possible to manufacture pellets with higher water contents from MX-80 raw material and production simulation runs could be made successfully for 6 mm and 8 mm sizes of bentonite pellet diameters. This means that it is possible to domestically produce in-house, customized extruded pellets for future research and actual operations, with the production rate of at 130 kg/h. The temperature gradients over the gap filling in the thermal conductivity tests varied between 2.0-4.1 °C/cm. The re-distribution of water content in the pellet filling was observed in all tests. The redistribution of water content was more significant with higher thermal gradient and longer testing time. The measured thermal conductivity of the roller compacted pellets was either 0.17 W/Km (dry) or 0.19 W/km (water containing). The used temperature gradient did not have a significant effect on the thermal conductivity values. The results of these experimental pellet studies are being used in the Finnish KBS-3V reference design work. Also the outcomes of the thermal conductivity task have been applied to long term safety related thermal modeling of the nuclear repository. The results are applicable to other waste management organizations and designs other than KBS-3V, where pellet filling of buffer and backfill areas is needed.

## INTRODUCTION

Finland's current reference design for high level radioactive waste disposal is based on the Swedish KBS-3V concept (Swedish abbreviation of Kärnbränslesäkerhet, Nuclear Fuel Safety). [1] In this design, vertical deposition holes are drilled in the bedrock to host spent nuclear fuel filled copper canisters embedded in bentonite clay. These vertical holes are made with certain manufacturing tolerances and also some extra clearance is needed for the actual bentonite buffer blocks installation process. Because of these facts there is always an unavoidable gap between the bedrock and compacted bentonite buffer. In the current Finnish vertical deposition hole buffer design, this gap has a nominal value of 50 mm with  $\pm 25$  mm tolerances and the gap is to be filled with bentonite pellets. The pellets are to be poured into the gap, without vibration or consolidation to achieve a bulk density of 1075 kg/m<sup>3</sup> [1]. Also the same kind of pellet filling material can be used to close the gaps between backfill blocks and the tunnel rock walls in the deposition tunnel section areas. The gap width between the backfill blocks and the theoretical tunnel wall/roof is designed to be 100 mm. The dry density of the backfill pellet fill shall be within the range of 900–1100 kg/m<sup>3</sup> [2].

The benefit of using gap filler is that it improves the properties of the components within the whole system especially with respect to thermal and mechanical transfer. Gap fillers can secure better

thermal conductivity and prevent spalling of the surrounding rock into the deposition hole. Also gap fillers might be needed to retain the adequate average density for example in cases when the gap itself might be enlarged or the density of the buffer blocks is lowered. Bentonite buffer blocks may need to be secured to stay in their places and also slowing down the possible water erosion (piping) may be needed for wet holes. The thermal conductivity of the pellet filling in the gap between the deposition hole rock surface and the bentonite buffer is one of the key parameters in thermal behavior of the bentonite buffer. The disposed copper canister produces residual heat due to decay of radioactive products. The decay heat is conducted through the bentonite buffer and the pellet filling to the surrounding rock mass. If the thermal conductivity of the pellet filling is too low the surface temperature of the canister will rise too high and this may lead for example to a case where the spacing between adjacent deposition holes should be increased.

This study focused on providing knowledge about bentonite pellets used for deposition of HLW with utilization in the KBS-3V concept. The main focus in the earlier studies was on the performance and properties of various commercially available bentonite pellets and granules [3]. The natural continuation step was to improve understanding on how to manufacture customized bentonite pellets for the chosen concept and how customized bentonite pellets perform in practice during the nuclear repository construction process [4, 5, 6, 7].

## METHODS

The bentonite material related studies presented in this paper mainly focused on laboratory experimental tests to optimize the pellet filling by customizing the raw materials and pellet manufacturing processes. Also the measurements of thermal properties of the pellets were emphasized. The bentonite pellets used in these studies could be divided to two main types by their manufacturing method. For the extrusion manufacturing type of bentonite pellets, both in-house made custom pellets and commercial ones were used. For roller compacted manufacturing method pellets, only commercial pellets were used which also acted as a reference type. All the bentonite pellets studied were intended for use in gaps between pre-compacted bentonite blocks and the rock walls in both buffer deposition holes and tunnel backfilling. Behavior and performance of the bentonite pellets were evaluated in the laboratory with selected tests. These included for example gap filling density, crush strength, free swelling and thermal conductivity.

## Extrusion Manufacturing of Bentonite Pellets

Twenty types of different extrusion pellets were manufactured at the VTT Technical Research Centre of Finland with laboratory and medium scale pelleting equipment. The main parameters that were varied during manufacturing included: bentonite raw material type, inert material additions, water content and pellet dimensions.

The extrusion manufacturing tests were done at facilities where presses are typically used for manufacturing biofuel wood–based pellets. The Amandus-Kahl 14-175 flat-die press setup was used in the first two extrusion test phases (Fig. 1.).



Fig.1. VTT's small pellet extrusion machine (left). Extrusion machine details: Pellet extrusion die (upper right) die and collar wheels (lower right).

The extrusion manufacturing tests were divided into three separate phases. In the first phase, a small extrusion pelleting machine was used and the main goal was to get operating staff familiar to work with bentonite material and to see if the pelleting by extrusion can be done with the available raw material compositions. Based on the initial results, more optimization studies were done with the same extrusion pelleting equipment. In the second phase, several different batches of bentonite pellets were done of a few kilos each. The scope of testing this time was to evaluate the pellet manufacturing process, with adjustments to parameters such as bentonite water content, bentonite feed rate to the extrusion machine, impact of cutting tools for controlling pellet length. The third and last phase was done with a medium-scale machine and had a main goal of producing over 100 kg batches of pellets having 17% water content of MX-80 bentonite raw material. This water content was the same as the current buffer reference design for buffer gap filling bentonite pellets [1]. Also to get 100 kg of pellet batch, the machine needed to be running continuously without overheating for some time (close to 60 minutes) which was also set as one of the secondary goals to simulate a full-scale production run. The summary of three extrusion tests is presented in Table I.

	Phase 1	Phase 2	Phase 3
Bentonite material	MX-80	Several*	MX-80
Water content	12-16%	16-21%	16-21%
Pellet diameter	7 mm & 8 mm	8 mm	6 mm & 8 mm
# Batches [kg]	12 [each 10 kg]	5 [each 5 kg]	5 [each > 100 kg]
* MX-80, 75%MX-80 +25% Illite, Milos B, Milos A, 75% MX-80+25% Sand			

TABLE I. Extrusion manufacturing test summary.

# **Gap Filling Tests**

The behavior of the customized bentonite pellets when filled into a gap was studied. This was investigated using the gap filling test arrangement developed at VTT and earlier studied with commercially available pellets [3]. The pellets to be studied were poured into a gap of 35 mm, with a surface face of 200 cm width by 114 cm height, using only the free falling technique without additional compaction or vibration. During the filling, the mass of each pellet batch was measured in order to get the total mass of poured pellets. After filling, first the pellet surface was leveled with a wooden mallet and then the pellet height was measured using digital images taken at various locations. For each pellet type the filling test was repeated three times. The schematic of the test arrangement is given in Figure 2. The gap filling test provided information on the bulk density achieved from the pellets after filling in the simulated deposition hole environment. Assessment considered aspects of compaction ability, achievable low to high range of poured bulk density, and handling considerations such as dust. Other details of the testing procedure followed the existing VTT methodology [3].



Fig. 2. The 35 mm gap filling test equipment filled with pellets.

#### **Thermal Conductivity Measurements**

Thermal conductivity values and measurement methods for bentonite pellet filling were also studied. The main objective of this task was to study how to measure thermal conductivity from larger sample sizes. Thermal conductivity of the bentonite pellets is depending on the water content, density and degree of saturation of the pellets, size and shape of the individual pellet and the geometry of the installation gap. The accuracy of the thermal conductivity measurement method is dependent upon the boundary conditions like the sample size in the test equipment.

Thermal conductivity of the pellet filling can be measured with a stationary heat flow where a constant thermal gradient is caused over the bentonite pellet filling. A typical stationary heat flow device for the thermal conductivity measurements is a heat flow meter which measures the density of heat flow rate through the specimen by a temperature difference generated by this density of heat flow rate crossing the specimen and the heat flow meter itself. The more detailed description of this method can be found from standard EN 12667:2001. During the tests the bentonite pellets were hand-compacted into an insulated frame 450-mm high, 450-mm, wide, and 80-mm depth (Fig. 3.). The frame was then inserted to the machine for measurements during 48 hours. For comparison purposes, measurements using the traditional thermal probe method were also taken. This method uses the transient technique with a small heated probe and it gives faster but often less accurate values.



Fig. 3. Thermal conductivity measurement of the bentonite pellets with heat flow meter method.

One goal of this work was to use even larger size of bentonite pellet batch than the heat flow meter would allow in a thermal conductivity study. To do this, a further development of the Hot Box (standards ISO 8990 (1994) and SFS-EN ISO 12567-1 (2000) device was commenced. The Hot Box consists of three chambers: the cold chamber, the warm chamber and the metering chamber that is located inside the warm chamber (Fig. 4.). The insulated metering chamber and the warm chamber to a minimum as the metering chamber and the warm chamber the warm chamber the same.



Fig. 4. Schematic arrangement of the Hot Box device.

A polycarbonate mold was built to hold the bentonite pellets (Fig. 5.). The outside dimensions of the mold were 1.19 m x 1.19 m and the width of the gap was 50 mm. The volume of the tested pellet filling was 70 liters. The thermal conductivity of the bentonite pellets was measured for two MX-80 raw material based bentonite pellet types: one was the roller compacted commercial pellets and the other was extruded in-house made pellets. The roller compacted pellets were pillow-shaped and the dimensions were  $12 \times 12 \times 6$  mm. The water content was 16.0% and the bulk density was 1075 kg/m<sup>3</sup>. One test was also carried out with oven dried pellets. The water content the extruded pellets was 17.0% and the bulk density was 986 kg/m<sup>3</sup>. These pellets were cylinder shaped and the diameter of the individual pellet was 8 mm. The pellets were installed into the mold by free-fall pouring. The temperature gradients over the gap filling material in the tests were chosen to be between 2.0–4.1 °C/cm. After the thermal conductivity tests were commissioned, samples were taken from the cold and warm sides of the pellet filling and the redistribution of the water content was determined.



Fig. 5. A polycarbonate bentonite pellet testing mold simulating a 50 mm gap.

# RESULTS

## **Extrusion Manufacturing**

The work done using extrusion pellets showed that it was possible to manufacture pellets with inert additions and with higher water contents, up to 21%, from MX-80. The extrusion tests also showed that the required production simulation runs could be made successfully with reference type of MX-80 bentonite raw material at 17% water content for 6 mm and 8 mm bentonite pellet diameters. This means that it is possible to domestically produce in-house customized extruded pellets for future research and actual operations.

Figure 6 shows the achieved production capacity of the different water content raw material and pellet diameter. The average production capacity for the medium size pelleting machine was found to be about 130 kg/h. There were not any issues when the medium size pellet production machine was used, though it needed some fine tuning to find the right parameters because the dimensions were different.



Fig. 6. Extrusion pellet production capacity of the medium size press.

## **Gap Filling Tests**

The gap filling tests were performed for two types of the in-house extrusion manufactured MX-80 pellets and roller compacted reference pellets. Pellets with additional installation method (spraying) and several other roller compacted pellets and one commercial extrusion pellet type are included for comparison purposes in the Figure 7. These results are shown as dry density values.



Fig. 7. Pellet dry density values from the gap filling tests.

#### **Thermal Conductivity Measurements**

Thermal conductivity of the reference roller compacted pellets was first measured with the thermal probe method. The bentonite pellets were hand compacted into the plastic cylinder and the calculated bulk density was 1045 kg/m<sup>3</sup>. The thermal conductivity tests were performed three times. The measured thermal conductivity values varied between 0.21–0.24 W/Km and the average thermal conductivity was 0.22 W/Km. The second set of thermal conductivity measurements were done using the heat flow meter method. The roller compacted bentonite pellets were hand compacted into an insulated frame and the calculated bulk density was 1257 kg/m<sup>3</sup>. The measured average thermal conductivity value was 0.19 W/Km.

The Hot Box measured thermal conductivities of the roller compacted pellets were 0.17 W/Km (dry) and 0.19 W/Km (water containing). The used temperature gradient did not have a significant effect on the thermal conductivity values. The thermal conductivity of the extruded pellets was 0.17 W/Km, which was 0.02 W/Km lower than with the roller compacted pellets at nearly the same water content.

The realized bulk densities were measured after the thermal conductivity tests. The bulk densities of the roller compacted pellets varied between 1085 to 1160 kg/m<sup>3</sup> and the bulk density of the extruded pellets was 1140 kg/m<sup>3</sup>. The measured bulk densities were larger than in the earlier investigations [3], where the widths of the gap were 25 mm and 35 mm.

## CONCLUSIONS

The work done using extrusion pellets showed that it was possible to manufacture pellets with higher water contents, with inert additions and production simulation runs could be made successfully for 6 mm and 8 mm sizes of bentonite pellet diameters. This means that it is possible to domestically produce in-house customized extruded pellets for future research and actual operations, with an achieved production rate of 130 kg/h. If the buffer design requires gap filling of a certain dry density value range of 820–1025 kg/m<sup>3</sup> to be used, this can be achieved with several pellets (both roller compacted and extruded) as shown in this study. Higher water content values allow closer compatibility with the designed bentonite buffer water content.

The temperature gradients over the gap filling in the thermal conductivity tests varied between 2.0–4.1 °C/cm. The redistribution of water content in the pellet filling was observed in all tests. The redistribution of water content was more significant with higher thermal gradient and longer testing time. The measured thermal conductivity of the roller compacted pellets was 0.17 W/Km (dry) or 0.19 W/Km (water containing). The lower value was measured with the dried bentonite pellets. The used temperature gradient did not have a significant effect on the thermal conductivity values. The thermal conductivity of the extruded pellets was 0.17 W/Km which was 0.02 W/Km lower than with the roller compacted pellets of nearly the same water content.

The results of these experimental pellet studies are being used in the Finnish KBS-3V reference design work. Also the outcomes of the thermal conductivity task have been applied to long term safety related thermal modeling of the nuclear repository. The results are applicable for other waste management organizations and designs other than KBS-3V, where pellet filling of buffer and backfill areas is needed.

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#### ACKNOWLEDGEMENTS

This work was commissioned by Posiva Oy, an expert organisation responsible for the final disposal of spent nuclear fuel of the owners. Posiva is owned by Teollisuuden Voima Oyj and Fortum Power & Heat Oy. Posiva is responsible for research into the final disposal of spent nuclear fuel of the owners as well as for the construction, operation and eventual decommissioning and dismantling of the final disposal facility.