A New Manufacturing Method of Bentonite Pellets as a Gap Filling Material for HLW Repository – 16110

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ABSTRACT
In high-level waste (HLW) repositories in Japan, there is a need to properly deal with gaps that surround buffers made of bentonite-based material, which are inevitably generated during construction. Bentonite pellets are thought to be a candidate material for filling these gaps. To fill the gaps densely in order to ensure desired buffer quality, high-density pellets with spherical shape would be suitable, and we have developed a new method for manufacturing such bentonite pellets, which is different from conventional methods. The new method first produces spherical pellets by extrusion and spheronization by alternately using two commercially available pelletizing machines. Then, the pellets, which do not have a very high density because of their water content, are turned into high-density ones using dry shrinkage. For typical Japanese sodium bentonite, we confirmed that spherical pellets can be manufactured when the water content is relatively high within the range of 25 to 30%. We observed that proper dry shrinkage enables the wet spherical pellets with a low density of about 1,500 kg/m^3 to turn into ones with a high dry density of about 2,000 kg/m^3. Drying slowly causes the proper dry shrinkage. We performed simple laboratory tests to evaluate the filling performance of pellets produced by the new manufacturing method. The maximum filling density obtained was 1,540 kg/m^3 when large (20-mm diameter) and small (1-mm diameter) pellets were mixed in the proportion of 60 % to 40 %, which is considerably higher than the previously reported maximum filling density in Japan, which was about 1,300 kg/m^3.

INTRODUCTION
In Japan, high-level waste (HLW) is being planned to be disposed of underground at depths exceeding 300 m, and it will be surrounded by a buffer made of bentonite-based material[1]. The buffer needs to have sufficient density to ensure low permeability. There are several ways of making buffers on site, and some methods generate gaps around the buffer, e.g., gaps between the buffer and host rock. These gaps must be properly dealt with in order to guarantee the effectiveness of the buffer.

Bentonite is considered to be a candidate material for filling these gaps because it has swelling characteristics and guarantees low permeability when used with a proper density[2]. As the gaps are generally narrow, the compacting of powdered bentonite in those areas not considered appropriate, unlike the case for making the buffer. Therefore, granulated bentonite, such as crushed bentonite and bentonite pellets, are expected to be effective materials for filling the gaps because they could achieve a decent density when they are simply poured into the gaps without vibration or tamping.

Previous studies on gap filling in Japan have examined several filling densities using various types of granulated bentonite. The filling materials employed are mainly crushed bentonite ores and commercially available bentonite pellets. For example, bentonite pellets are made using roller compaction and briquetting techniques, and they have been used in other countries to investigate filling techniques[3,4]. Previously the maximum filling density that have been achieved in Japan when filling gaps was about 1,300 kg/m^3. Because the dry density of buffers in Japan is 1,600kg/m^3 for a mixture of bentonite (70%) and sand (30%), if the gap is to have the same functionality as the buffer, a filling density of at least 1,370 kg/m^3 is preferred as the dry density when the pellets comprise only bentonite, and contain no sand or other materials.

Pellets manufactured by various methods have different properties such as density, size, and shape, which results in different filling densities. It is not fully known which of the properties of bentonite pellets contribute to the increase of the filling density, but we considered that high-density pellets are effective, and that the spherical shape is suitable for pouring.

As the applied compressive force increase, we obtain a higher density of bentonite pellets. However, no commercially available pelletizing machines can apply such high compression forces to realize higher pellet densities, and to achieve higher compression, we require the use of large-scale and costly machines. It is also difficult to make spherical pellets under high compression.

In this study, we developed a new method for the manufacture of high-density spherical bentonite pellets. The proposed method uses the dry shrinkage mechanism instead of mechanical compression to obtain higher pellet density. First, we explain the procedure of the method and machines that were used. Next, we present the results of some manufacturing tests that were conducted to determine the manufacturing conditions. Finally, we performed simple laboratory tests to evaluate the filling performance of the pellets that were manufactured using the new method .

NEW MANUFACTURING METHOD

In this paper, we propose a new method for manufacturing spherical, high-density bentonite pellets. The method is divided into two processes, namely the pelletizing and drying processes. In the pelletizing process, we first manufacture the cylindrical pellets using a pelletizing machine called an extruder and they are then converted into spherical pellets using another pelletizing machine called a spheronizer. Both the extruder and spheronizer are commercially available.

The spherical pellets that are manufactured in the pelletizing process have a relatively high water content, resulting in a not-so-high density of about 1,500 kg/m^3. Even pellets with such low-densities can be converted into pellets with a high density of about 2,000 kg/m^3 if they are dried under proper conditions.

Pelletizing Process

Extruders are commercially available pelletizing machines that produce cylindrical pellets by passing moderate wet material through holes in a steel disk called a die. As shown in Fig. 1, water-controlled material is supplied on the die. Then the roller crushes the material on the die, and the material is subsequently extruded through the holes. Using a die having holes with different diameters, as shown in Fig. 2, we obtain extruded materials with different diameters. The extruded materials are cut into appropriate lengths by placing a cutter under the die, so they become cylindrical bentonite pellets, as shown in Fig. 3.

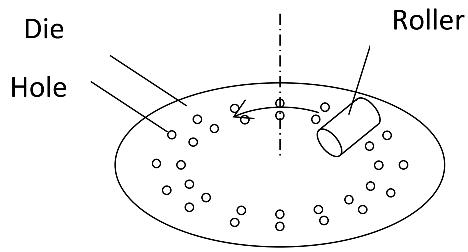


Fig. 1. Conceptual view of extruder

Fig. 2. Steel disks called die on which many holes are arranged. The left image is an 8-mm die with 8-mm-diameter holes and the right image is a 4-mm die with 4-mm-diameter holes.

Fig. 3. Cylindrical pellets after extrusion using an 8-mm die

Spheronizers are different pelletizing machines that convert the cylindrical pellets produced by extruders into spherical pellets. The cylindrical pellets supplied on the spinning disk are made to collide with the wall using the centrifugal force (Fig. 4). The complicated action involves the collision and the friction between pellets causes the cylindrical pellets to become rounded, resulting in spherical pellets. The appearance of successfully pelletized pellets is nearly spherical, and have a black surface, as shown in Fig. 5.

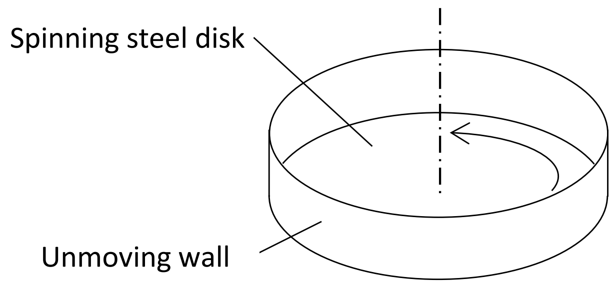


Fig. 4. Conceptual view of spheronizer

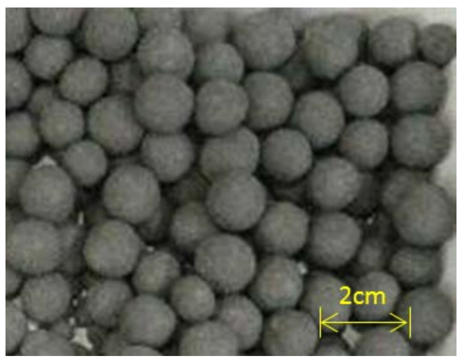


Fig. 5. Appearance of spherical pellets after spheronization. Examples of pellets made by extrusion using an 8-mm die.

Drying Process

The drying process plays a critical role in increasing the density of after-pelletizing spherical pellets. In this process, the inducing of dry shrinkage on the pellets leads to an increase in the dry density.

MANUFACTURING TESTS

Manufacturing tests consist of pelletizing tests and drying tests. Pelletizing tests confirm the possibility of obtaining spherical pellets from bentonite and an appropriate water content, while drying tests reveal the density-increasing effect on dry shrinkage, and the dry condition for manufacturing high-density bentonite pellets.

Pelletizing Test and Water Content

To produce spherical pellets using both of the pelletizing machines, the water content is very important for both extrusion and spheronization. During extrusion, a water content that is too large prevents the material from being converted into a cylindrical shape, while a water content that is too small causes it to become struck in the hole and no extrusion is achieved. Similarly, during spheronization, if the water content of the cylindrical pellets increases, they stick to each other and become large irregular chunks. On the other hand, if the water content decreases, the cylindrical pellets become an ellipsoidal or dumbbell-like shaped but they do not have spherical shapes.

This method is typically used in the manufacture of fertilizers and agrochemicals. In these fields, bentonite is sometimes used as a material for pelletizing, not as the primary material but as an additive. Bentonite is a very sticky material, and it is therefore used as a binder when manufacturing fertilizers and agrochemicals. There have been no reports of the manufacture of bentonite-only pellets, whether in the radioactive waste disposal field or in other fields. Hence, it is not clear whether bentonite solely can be manufactured into spherical pellets.

We conducted pelletizing tests to confirm whether proper water content is exist or not, and, if exist, how much the proper water content is for manufacturing. For the bentonite, we used Kunigel V1, which was supplied by Kunimine Industries Co., Ltd. It is a commercially available typical sodium-type bentonite. As the main machines, we used a Disk Pelleter model F-20 as an extruder and a Marumerizer model Q-400 as a spheronizer, both of which were supplied by Dalton Corporation in Japan.

For the tests, we used dies with holes having five different diameters. The hole diameters were 8 mm, 5 mm, 4 mm, 2 mm, and 1 mm. First, bentonite with a 28.95% water content was extruded using the 8-mm die, and it was then spheronized. We determined the setup water content based on the experience of engineers. More specifically, the amount of water added to the bentonite was gradually varied, and we verified the condition of the mixture by hand. We successfully manufactured spherical pellets using an 8-mm die. For the 5-mm die, we set the same water content of 28.95%, and the pellets were successfully manufactured. For the 4-mm die, two water contents were specified. Pellets with a water content of 27.48% were successfully manufactured, but the cylindrical pellets with a 25.32% water content made by extrusion could not be converted into a spherical shape. For the 2-mm die, we used a water content of 27.06% because we evaluated by our experiences that a lower water content was suitable for the die with holes having a diameter smaller than a 4 mm. We obtain successful results. Finally, we applied to the 1-mm die bentonite with two water content values. In the case of the higher water content of 27.06%, the cylindrical pellets stuck to each other and became large irregular chunks during spheronizing, and no spherical pellets were obtained. In contrast, for the case with a water content of 25.98%, the cylindrical pellets were successfully converted into a spherical shape.

The results in all of the cases were combined in Fig. 6, where black circles show conditions that are suitable for pelletizing, and cross marks show those that are unsuitable. We confirmed the existence of suitable water contents for bentonite pelletizing. To manufacture spherical bentonite pellets, the water content of bentonite before pelletizing should be roughly 25% to 30%, which is relatively high compared to that for buffer production with compaction. The adequate water content varies according to the extrusion diameter. When pellets with larger diameters need to be manufactured, a slightly higher water content should be adopted.

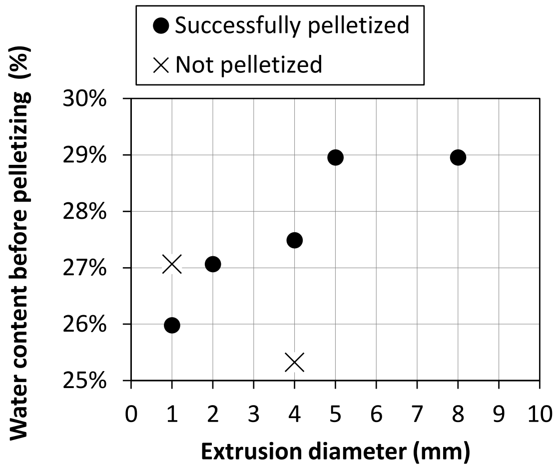


Fig. 6. Results of the pelletizing tests, indicating suitable water content values.

Drying Tests

The water content of spherical pellets after the pelletizing process was 25 to 30 %. Because the color was closer to black than to grey (Fig. 5), the pellets can be considered to have been in a nearly saturated condition. Therefore, the dry density of the pellets is assumed to be about 1,500kg/m^3, which is lower than that of other bentonite pellets that were manufactured using compaction-based methods. However, dry shrinkage after pelletizing may contribute to an increase in the dry density of the pellets.

To confirm the effect of dry shrinkage on increase in the density, we conducted drying tests under different drying conditions. We used two types of drying equipment in the tests. One is a dry oven with a blow that is generally used to dry various pellets, and the other is a dry oven without a blow. Dry temperatures were low (40 or 50°C.) and high (110°C). For the tests, we used spheronized pellets made by extrusion employing 5-mm and 8-mm dies. We measured changes in water content at suitable intervals during the drying. The dry densities of the pellets were measured after completing the drying.

Fig. 7 and Fig. 8 show the changes in the water content obtained using the two types of ovens with different temperatures. Using the blow-oven (Fig. 7), the pellets dried quickly, while the pellets dried slowly using the non-blow-oven (Fig. 8(a) and (b)). We see that the ambient air of the pellets in the non-blow-oven increased in humidity during drying, preventing water evaporation from the pellets.

The dry densities of after-dried pellets are shown in Fig. 9. We found that the drying method affects the dry densities of the pellets after drying. The pellet density reached its highest level when the pellets were dried by the non-blow-oven at low temperatures. Thus, it could be argued that a low drying rate helps low-density pellets to obtain better dry shrinkage, and turn into high-density pellets. A suitable dry shrinkage enabled low-density bentonite pellets to be converted into high-density ones with a dry density of about 2,000 kg/m^3.

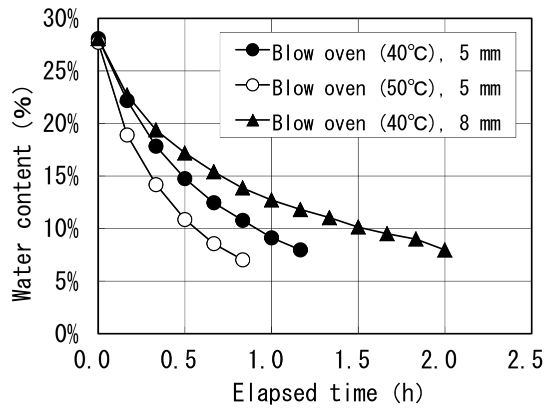
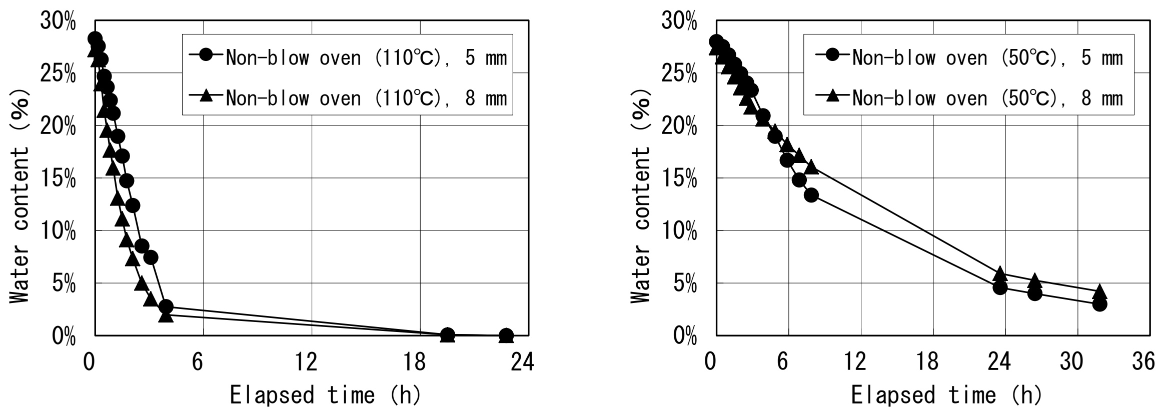


Fig. 7. Changes in water content during drying with blow-dry-oven.



(a) at high temperature (b) at low temperature

Fig. 8. Changes in water content during drying with non-blow-dry-oven.

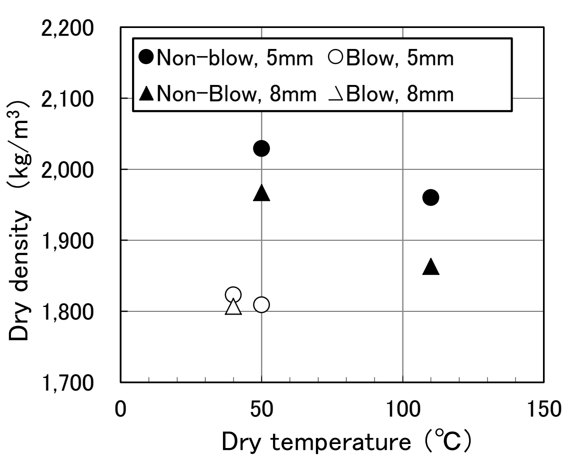


Fig. 9. Difference in dry density of dried pellets corresponding to drying manner and temperature.

FEATURES OF BENTONITE PELLETS MANUFACTURED USING PROPOSED METHOD

Fig. 10 shows the grain-size distribution of actual pellets that were previously manufactured using the newly proposed method described above. We manufactured pellets with a wide range of sizes. The new method enables relatively simple manufacturing of bentonite pellets with different sizes by only switching a die to another one that has holes of a different diameter. Two other features of pellets regarding to their size are: each pellets has a low variation in the grain size, and the actual grain size is slightly larger than the extrusion diameter. Fig. 11 shows examples of completed bentonite pellets. The pellets, which were withdrawn from the middle of grain distribution, appear to have an extremely spherical shape.

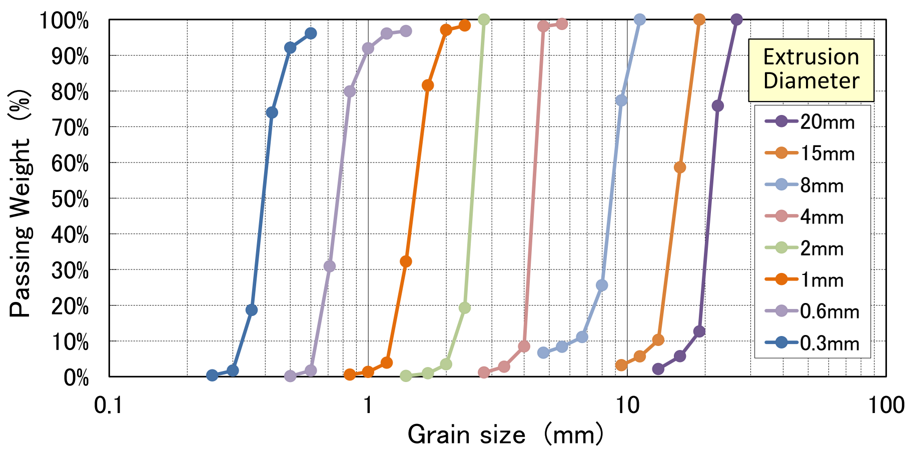


Fig. 10. Size distribution of pellets manufactured by the new manufacturing method.

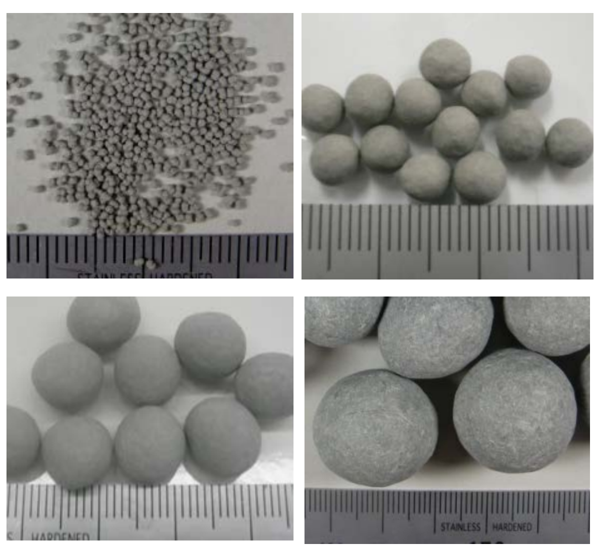


Fig. 11. High-density spherical pellets manufactured using the method described in this paper. Extrusion diameters are 1 mm, 4 mm, 8 mm, and 20 mm.

FILLING TEST

We performed laboratory filling tests to evaluate the gap filling performance of the pellet produced using the proposed manufacturing method. We measured the filling performance in terms of the filling density, which is given by the ratio of the dry mass of the pellets filled into a container simulating a gap to the container volume.

Pellets and Gap-simulating Container Used for Filling Tests

In the test, we used five types of pellets with different sizes. The extrusion diameters of the pellets were 1 mm, 2 mm, 4 mm, 15 mm, and 20 mm. Prior to the filling test, all pellets were oven-dried at 110°C for at least 24 hours. We assumed that the drying process would not promote dry shrinkage any further because all of the pellets had a water content of less than 10%. The dry densities of the pellets were measured in advance based on Archimedes' principle to be sunk in water (Table I)

The gap was simulated using a small cylindrical container with an inner diameter of 100 mm and a depth of 127 mm, giving a volume of 1,000 cubic centimeters (Fig. 11).

Test Cases and Filling Procedure

When spherical pellets with the same diameter are used for filling, only low filling density is obtained. In fact, when only one type of pellet was individually poured into the cylindrical container, we obtained a filling density as low as 1,310 kg/m^3 at the maximum, as shown in Table I, although the pellet density was as high as about 2,000 kg/m^3.

We expected that a mixture of two types of pellets having different sizes would be effective for increasing the filling density. Then, we conducted filling tests by varying both the pairing of different size pellets and the mixing rate.

A filling procedure was used in which two types of pellets were poured separately in five nearly equal layers of the container. Other operations such as tamping and vibration were not employed. Fig. 12 shows an example of the progress of the filling test.

Results

Fig. 13 shows the results of the filling tests. Mixtures of different-size pellets filled the container more densely than non-mixed pellets. The mixing rate significantly affected the filling density. The maximum filling density was obtained when the mixing rates ranged from 40% to 50% for all pairings. The influence of pairing was also strong, and a combination of the largest and smallest pellets was more effective for achieving the highest filling density.

We obtained a maximum filling density of 1,540 kg/m^3 under the condition where 20-mm pellets (60%) and 1-mm pellets (40%) were mixed. The maximum filling density obtained was considerably higher than any filling density previously reported, the maximum of which was about 1,300 kg/m^3.

TABLE I. Pellet density and filling density using individual pellets.

Pellets type	Dry density of pellets (kg/m^3)	Filling density (kg/m^3)
1 mm pellets	2.16	1.31
2 mm pellets	2.10	1.26
4 mm pellets	2.04	1.21
15 mm pellets	1.97	1.12
20 mm pellets	2.00	1.11

(a) Fourth layer completed.

(b) Large pellets poured on the completed fourth layer

(c) Fifth layer completed after pouring small pellets

Fig. 12. Filling test in progress (15-mm and 2-mm pellets with a mixing ratio of 60%:40% in dry weight).

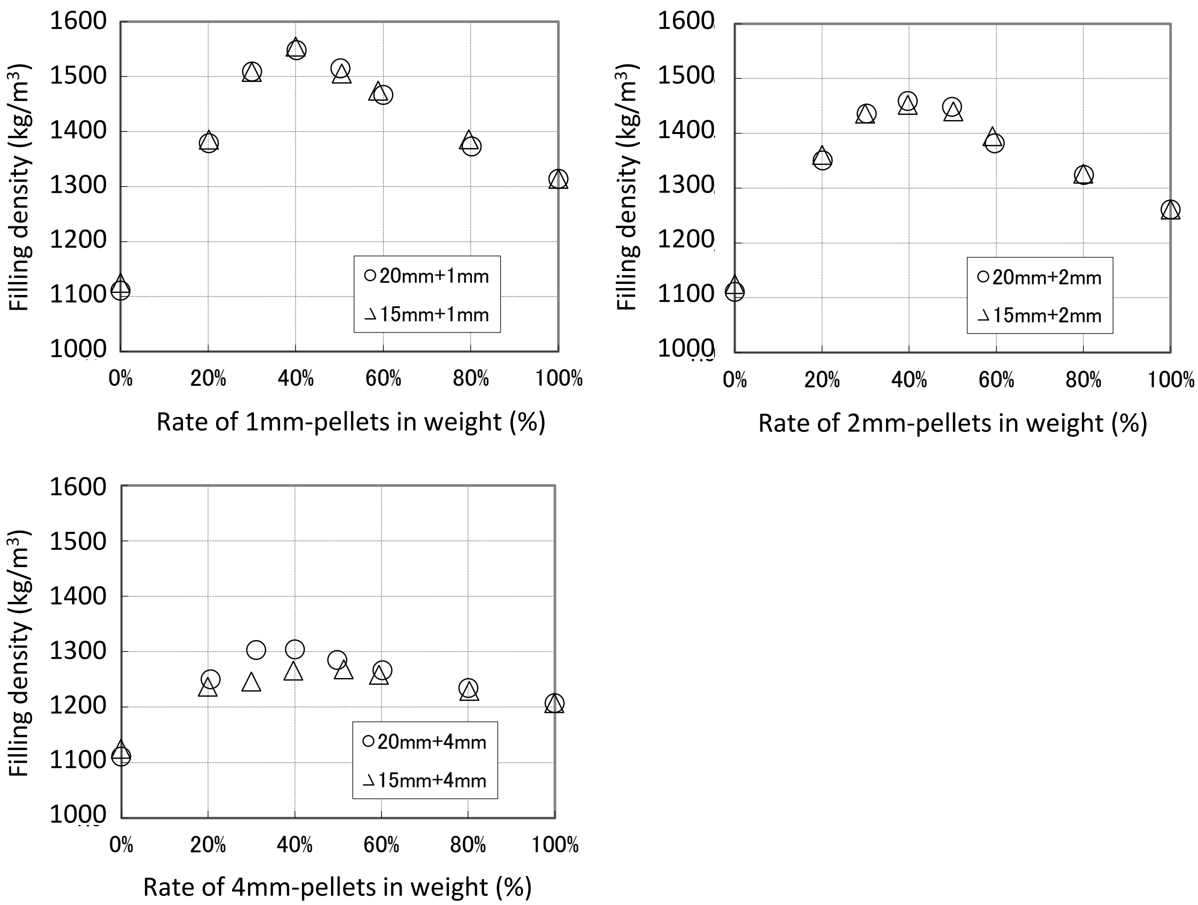


Fig. 13. Filling density of combination of large and small pellets for varying mixing rate.

CONCLUSIONS

In this study, we proposed a new method for the manufacture of bentonite pellets as the material for filling gaps around a buffer made of bentonite. By conducting manufacturing tests, we confirmed that high-density spherical pellets could be manufactured under suitable conditions. A feature of the method is the active use of dry shrinkage for enhancing pellet density instead of the application of compressive force. Dry shrinkage converts low-density spherical pellets into high-density ones, where the low-density pellets have relatively high water content, and were produced using general pelletizing machines

Pelletizing tests, which comprised extrusion and spheronizing, indicated the need for suitable water content of raw materials for producing spherical pellets. The water content ranged roughly from 25% to 30%, which is slightly different from that of a different pellet size.

Drying tests, which is another manufacturing test, indicated the need for suitable drying conditions for enhancing the pellet density. It was shown that appropriate dry shrinkage could enable the conversion of low-density bentonite pellets into high-density ones with a dry density of about 2,000 kg/m^3.

We conducted filling tests to confirm the filling performance of the pellets that were manufactured using the newly proposed method. Pellets have a feature of narrow grain-size distributions, then filling density is not very high when a single type of pellet is used. However, a combination of two different size pellets achieves a higher filling density. When larger pellets and smaller pellets are mixed with a ratio of about 50:50, the filling density becomes a maximum. In this study, by mixing 20-mm and 1-mm pellets in a 60:40 ratio, we achieved a filling density of 1,540 kg/m^3, although only simple laboratory tests were conducted.

In the future, we will investigate the development of full-size filling equipment, we will perform full-scale experiments and studies on the behavior of filled bentonite pellets.

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