Effect of granule size distribution on the compaction property of GMZ bentonite

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Abstract. In the current concept for disposal of high-level radioactive waste (HLW) in China, a barrier composed of blocks of highly compacted bentonite is constructed around the waste canisters. To develop this technique, uniaxial compaction of full-sized bentonite blocks has been initiated. Many factors affect the quality of bentonite blocks, including water ratio and granule size distribution of the bentonite, compaction pressure and rate, and form geometry. In the present study, compaction tests were conducted using powdered, pelleted, and mixed GMZ bentonite to evaluate the effects of granule size distribution on the integrity and uniformity of the block samples. Results show that high-quality blocks can be manufactured by adjusting the water content of the bentonite to 17%, in spite of different granule size distributions. The compaction in the mixed bentonite was better than that in the powdered and pelleted samples. However, the difference in final density was negligible when compacting at the same pressure. The block uniformity was not improved by adopting different granule size distributions when the height:diameter ratio of the bentonite samples reached 2. Blocks produced by powdered and mixed bentonite were prone to radial cracks when subjected to moisture loss; however, the cracks were healed after moisture redistribution.

1. Introduction
Geological disposal is the preferred option for the final disposal of high-level radioactive waste (HLW) in most countries. Moreover, nearly all treatment programs for HLW are considering the use of engineered barriers\[1\]. Because of its low hydraulic conductivity, high swelling capacity, and good adsorption properties, Gaomiaozi (GMZ) bentonite has been proposed as a suitable buffer/backfill material for the construction of deep geological repositories in China\[2\]. To meet the required safety functions of buffer/backfill material, the raw bentonite material must be compactable to a relatively high density. Two main techniques are used for compacting blocks of bentonite: isostatic compaction and uniaxial compaction\[3\]. Because the uniaxial compaction technique requires much less preparation and the blocks do not require reshaping after compaction\[4\], this method has been intensively investigated\[5, 6, 7\]. Many factors, including water ratio and granule size distribution of the bentonite, compaction pressure and rate, and form geometry, affect the quality of bentonite blocks\[8, 9\]. However, the effects of the granule size distribution on the quality and compaction properties of bentonite blocks remain poorly understood. It is believed that fine powder does not facilitate the escape of trapped air during the compaction process and may lead to crack generation in the blocks. Moreover, the coarser granules contained in bentonite pellets lead to brittle edges, and the strength of the blocks can reduce to the extent that they cannot be safely lifted\[10\].

In this study, compaction tests were conducted using powdered, pelleted, and mixed GMZ bentonite to evaluate the effects of granule size distribution on the integrity and uniformity of bentonite blocks.
2. Material and methods

2.1. Materials

GMZ bentonite pellets obtained from Gaomiaozi County, Inner Mongolia Autonomous Region, China were used to prepare materials with different granule size distributions. The pellets were composed of bentonite particles disintegrated from bentonite ore during the air-drying process; particles larger than 4 mm and smaller than 0.9 mm were removed using a vibrating screening machine. As shown in the basic mineralogical composition of the bentonite pellets summarized in Table 1, the average montmorillonite content was 47.8%. The cation exchange capacity of the bentonite pellets was 58.3 mmol/100 g, and the main exchangeable cations were Na$^+$ (28.5 mmol/100 g), Ca$^{2+}$ (17.2 mmol/100 g), and Mg$^{2+}$ (10.6 mmol/100 g). The bentonite powder was prepared by milling the bentonite pellets and screening with a standard 50 sieve. The mixed bentonite was obtained by mixing bentonite pellets and powder in a 3:2 proportion by dry weight.

| Sample No. | Montmorillonite | Quartz | Cristobalite | Feldspar | Zeolite | Mica |
|------------|-----------------|--------|--------------|----------|---------|------|
| 1          | 44.6            | 26.2   | 0.8          | 18.6     | 8.6     | 1.2  |
| 2          | 49.8            | 25.8   | 1.1          | 15.6     | 6.2     | 1.5  |
| 3          | 48.9            | 26.9   | 0.9          | 15.5     | 7.0     | 0.8  |

The granule size distributions of the pelleted, powdered, and mixed bentonite were determined by sieving using standard sieves. To avoid crushing of the granules, sieving was performed manually rather than using a vibration machine. The granule size distribution results are presented in figure 1. The granule size of the bentonite powder was less than 0.3 mm, among which the content of soil particles less than 0.1 mm was about 85%. Of the bentonite pellets, 90% were 1–2 mm in diameter. Particles larger than 0.1 mm and smaller than 0.1 mm accounted for about 50% of the mixed bentonite, and the content of particles larger than 1 mm was about 40%.

![Figure 1. Granule size distribution of the investigated bentonite materials](image_url)

2.2. Test procedures

To investigate the compaction characteristics of the bentonite materials with different granule size distributions, cylindrical samples with weight $m = 206.8$ g and diameter $\varphi = 50$mm were uniaxially compacted to 100 MPa at a rate of 2 mm/min using a rigid stainless steel mold on a 200-T hydraulic press, with the maximum load kept for 10 minutes. The compaction forces and displacement of the
samples were recorded continuously. The weights and dimensions of the samples were measured after they were ejected from the mold, and their dry density was calculated.

In the sample uniformity test, the same amounts of raw materials with different granule size distributions were compacted to obtain φ50 mm × 100 mm samples with the goal of reaching the same dry density of 1.75 g/cm³. After being ejected from the mold, the samples were cut into five φ50 mm × 20 mm slices using a band saw. Each slice was then cut into two pieces. One piece was dried in a 105°C oven for 24 hours to determine the water content, whereas the other was submerged in paraffin oil with known density to measure the bulk density of the slice.

A relative humidity-induced cracking test was conducted to evaluate how the compacted bentonite blocks with different granule size distributions react when exposed to different relative humidity levels. The influence of granule size distribution on the healing ability of the cracks was also investigated. All specimens were compacted using 82.6 g of material on a 200-T hydraulic press with a target dry density of 1.75 g/cm³. The sample dimensions were φ50 mm × 20 mm. After manufacturing, the samples were vacuum packed in plastic bags and stored in self-sealed bags to prevent drying. Figure 2 shows the prepared samples. After all samples were prepared, a climate chamber was used to set the storage temperature to 25°C, and the humidity was set to 30%, 50%, 75%, and 95%. The weight of the samples was measured regularly, and the generation and development of cracks were recorded by taking photographs. The weighing and photographing intervals were once every 2 hours during the first 10 hours, twice daily on the second and third days, and once daily during the remaining test time.

3. Results and analysis

3.1. Effect of granule size distribution on compaction property

The compaction forces and displacement of the samples were continuously recorded during the compaction property test. The corresponding displacement under different stress levels was determined from the stress–displacement curve to calculate the instantaneous dry density of the samples during the compaction process. Figure 3 shows the correlation between compaction pressure and instantaneous dry density. When the dry density was lower than 1.9 g/cm³, the required compaction pressure for the three types of bentonite showed negligible difference. When the dry density was greater than 1.9 g/cm³, the required compaction pressure increased rapidly, and obvious differences in the compaction property appeared among the three materials. The pelleted bentonite was the most difficult to compact, with a maximum instantaneous dry density of 1.94 g/cm³, while that of powder bentonite was 1.97 g/cm³. Moreover, the mixed bentonite compacted relatively easily and had a maximum instantaneous dry density of 1.99 g/cm³.

Figure 4 shows photographs of the bentonite samples composed of the three types of bentonites. No obvious cracking or dropped pieces were observed, and the quality of all the samples was judged to be good.
Table 2 gives the height and diameter of the samples measured before compaction and after ejection from the mold. The filling factor of the three types of bentonites at 100 MPa, i.e., the ratio of the height of the test sample after filling into the mold to the height after compaction, differed significantly. The pelleted bentonite had the largest filling factor of 1.94, followed by powdered bentonite at 1.86, while the mixed bentonite had the smallest value at 1.69. As reported in previous research, raw materials with a smaller filling factor have obvious advantages in reducing the height of the mold, improving compaction efficiency, and reducing block manufacturing cost [11]. Although the instantaneous dry density of the three types of bentonites can reach more than 1.9 g/cm³ in the process of compaction, the rebound phenomenon of bentonite blocks during and after the unloading of pressure causes the dry density of the samples to reduce to 1.81–1.84 g/cm³. The dry density was lowest in the pelleted bentonite and highest in the mixed bentonite.

![Figure 3. Correlation between compaction pressure and instantaneous dry density (φ50 mm × 50 mm)](image)

![Figure 4. Photographs of bentonite blocks (50 mm × 50 mm) composed of bentonite materials with different granule size distributions](image)
Table 2. Parameters of bentonite blocks (φ50mm × 50mm) composed of materials with different granule size distributions

| Bentonite type | Height before compaction | Moisture content | Height after expulsion | Diameter after expulsion | Dry density after expulsion |
|---------------|--------------------------|------------------|------------------------|--------------------------|---------------------------|
|               | mm                       | %                | mm                     | mm                       | g/cm³                      |
| Powder        | 91.35                    | 16.83            | 49.00                  | 50.08                    | 1.83                      |
| Pellets       | 95.50                    | 17.19            | 49.35                  | 50.09                    | 1.81                      |
| Mixed         | 82.24                    | 17.26            | 48.60                  | 50.09                    | 1.84                      |

3.2. Effect of granule size distribution on block uniformity

Figure 5 shows the dry density distribution along the height direction of φ50 mm × 100 mm samples containing the three different bentonite types. The dry density of each 20-mm slice of mixed bentonite was higher than that of the other two types in the same location; in particular, the mixed bentonite was easier to compact under the same conditions. However, the heterogeneity of the samples from top to bottom was essentially the same. This indicates that when the height:diameter ratio reaches 2, even if the moisture content of the raw materials is adjusted to 17%, which is a relatively high water ratio used to reduce the friction between particles as well as that between the particles and the wall of the mold, adopting different granule size distributions is unlikely to make the blocks more uniform.

![Figure 5](image)

Figure 5. Dry density as a function of the distance from the top surface of the samples after compaction (φ50 mm × 100 mm)

3.3. Crack generation and development in different humidity environments

Figure 6 shows the weight change of the samples in different humidity environments. The change in sample weight began immediately after the samples were placed in the climate chamber. After about 50 hours, the weight change tended to be moderate. Regarding the three types of bentonite samples with the water content adjusted to about 17%, all samples lost weight under relative humidity levels of 30%, 50%, and 75%. In particular, weight loss was more than 8% under relative humidity levels of 30% and 50%. Under 95% relative humidity, the three types of bentonite lost slight amounts of water after being placed in the environment and then slowly absorbed the water. However, weight change was within ±0.6%. Under the same relative humidity, the weight changes in the pellets, powder, and mixed bentonite were essentially the same. No obvious difference was noted under 95% relative humidity; however, the mass loss of the mixed bentonite was slightly lower than that of pellets and powder under relative humidity levels of 30%, 50%, and 75%.

![Figure 6](image)
Figure 6. Changes in the mass of bentonite block samples with different granule size distributions

Periodically taking pictures of the samples under the four humidity environments enabled the observation of the occurrence and development of cracks on the sample surfaces. Because of the relatively small change in mass, no cracking was observed in the samples of the three different materials under 95% relative humidity. Moreover, no cracking was recorded in compacted samples of pelletized bentonite under the four humidity conditions, although the mass changes were significant under relative humidity levels of 30% and 50%. However, radial cracks appeared on the surfaces of compacted samples of powdery and mixed bentonite (Figure 7) after exposure in the climate chamber for 2 hours at relative humidity levels of 30%, 50%, and 75%. The cracks tended to be obvious as the humidity decreased; in particular, the amount of water loss increased. Under the same environment, cracks in powdery bentonite samples were more obvious than in mixed bentonite. After 24–48 hours of testing, all the cracks were healed and then disappeared (Figure 8). By the end of the test, no new cracks were observed.
Figure 7. Surface cracks in various samples under 30% relative humidity

Figure 8. Crack healing in bentonite powder under 30% relative humidity

The relative humidity-induced cracking tests showed that bentonite of different granule sizes with water content adjusted to about 17% lost water under relative humidity levels of 30%, 50%, and 75%. The lower the humidity, the more the amount of water lost. At a relative humidity of 95% essentially no change in weight was noted. The pelleted bentonite samples did not produce cracks under the four humidity environments, whereas the powdered and mixed bentonite samples exhibited radial cracks under relative humidity levels of 30%, 50%, and 75%. The lower the humidity, the more obvious the cracks. Although the cracks in the powdered bentonite samples were more pronounced than those in the mixed bentonite samples, under this test condition, the samples did not collapse and could be handled in a safe manner. After a certain period of time, the cracks self-healed and disappeared. Therefore, the sensitivities of the investigated materials with different granule sizes to humidity had negligible differences.

4. Conclusions
Compaction tests of three bentonite materials with different granule size distributions and maintenance tests under different humidity conditions were conducted to investigate the effects of granule sizes on the quality and integrity of bentonite blocks. The conclusions of this study are as follows:
(1) Irrespective of the form of the raw materials of GMZ bentonite, i.e., pellets or powder, or whether the two are mixed in a certain ratio to obtain materials with a specific granule size distribution, high-quality blocks can be manufactured when the moisture content is adjusted to about 17%.
(2) Bentonite in the form of fine powder and coarse granules has better compaction properties than that containing only fine powder or large pellets. Mixed bentonite has a lower filling factor, which is advantageous for reducing the height of the mold and improving the compaction efficiency.
(3) The granule size distribution has a negligible effect on improving the density gradient along the height of a cylindrical bentonite block.
(4) Blocks composed of pelleted bentonite are not prone to crack generation, whereas those composed of powder and mixed bentonite produce radial cracks when subjected to significant moisture loss. However, these cracks can self-heal and disappear. Therefore, the difference in sensitivity of the investigated materials with different granule sizes to humidity is negligible.

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