

PHYSICAL PROPERTY OF CHINA'S BUFFER MATERIAL FOR HIGH-LEVEL RADIOACTIVE WASTE REPOSITORIES

WEN Zhijian

(Beijing Research Institute of Uranium Geology, China National Nuclear Corporation, Beijing 100029, China)

Abstract: The deep geological disposal is regarded as the most reasonable and effective way to safely dispose high-level radioactive wastes(HLW) in the world. The conceptual model of HLW geological disposal in China is based on a multi-barrier system that combines an isolating geological environment with an engineered barrier system including the vitrified HLW, canister, overpack and buffer/backfill material. The bentonite is selected as base material of the buffer/backfill material in HLW repositories, due to the very low permeability and excellent retardation of nuclides from migration, etc. GMZ deposit is selected as the candidate supplier for buffer material of HLW repositories in China. Since 2000, systematic study was conducted on GMZ - 1 that is Na-bentonite produced from GMZ deposit and selected as reference material for Chinese buffer material study. The mineral composition, basic parameters of GMZ - 1 bentonite and thermal conductivity, hydraulic conductivity, unconfined compression strength as function of dry density and water content are presented. The swelling stress of GMZ - 1 bentonite as function of dry density is also reported. GMZ - 1 bentonite is characterized by high content of montmorillonite(about 75%) and less impurities. The adequacy understanding of property and long-term behavior in deep geological condition of GMZ - 1 is essential to safe dispose the high-level radioactive wastes in China.

Key words: high-level radioactive wastes disposal; GMZ - 1; buffer material; physical properties

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中国高放废物处置库缓冲材料物理性能

温志坚

(核工业北京地质研究院, 北京 100029)

摘要: 深地质处置被国际上公认为处置高放废物的最有效可行的方法。中国深地质处置的概念模型采用多重工程屏障系统(包括废物固化体、废物容器、外包装、缓冲/回填材料)和适宜的地质围岩地质体共同作用来确保高放废物与生物圈的安全隔离。膨润土由于具有极低的渗透性和优良的核素吸附等性能而被国际上选作缓冲材料的基础材料。经过全国筛选,高庙子膨润土矿床被选作我国缓冲材料供应基地。从2000年起,对产自该矿床的钠基膨润土 GMZ - 1 开始了系统的研究工作。介绍了 GMZ - 1 的矿物组成、基本特征和 GMZ - 1 在不同干密度、不同含水量条件下的热传导、水传导、力学性能参数及 GMZ - 1 在不同干密度条件下的膨胀特性参数测定结果。GMZ - 1

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Corresponding author: WEN Zhijian(1968 -), male, Ph. D., obtained his bachelor degree at Department of Geology, Peking University, Beijing in 1990. He is presently the professor at Research Center for Environmental Protection, Beijing Research Institute of Uranium Geology. His main research activities cover nuclear waste disposal and environmental remediation. E-mail: wenzhijian@hotmail.com

钠基膨润土具有蒙脱石含量高(75%左右)、杂质矿物相对较少的特点,对于该材料的系统和深入研究对于开发我国缓冲回填材料技术,确保高放废物的安全有效处置有重要意义。

关键词: 高放废物处置;高庙子膨润土;缓冲材料;物理性能

1 INTRODUCTION

Radioactive waste is produced from a wide range of human activities. The wastes are in many different physical and chemical forms, contaminated with varying activities of radionuclides. High-level radioactive waste(HLW) is characterized with very high initial radioactivity, and presents a potential long-term risk, even though it decreases with time, Total activity is initially dominated by beta/gamma emitting short-lived fission products that decay considerably over several decades to hundreds years. Over a longer timescale, the longer-lived actinides and their radioactive daughters(many of them are alpha-emitters) make a more significant contribution to total activity. The safe disposal of high-level radioactive waste is a key requirement of the nuclear industry worldwide.

Among the options discussed for disposing of HLW, an international consensus has emerged that deep geological disposal on land is the most appropriate means for isolating such wastes permanently from environment^[1].

The concept of geological disposal of high-level radioactive wastes in China is based on a multi-barrier system that combines an isolating geological environment with the vitrified HLW, canister, overpack and buffer/backfill material(Fig.1).

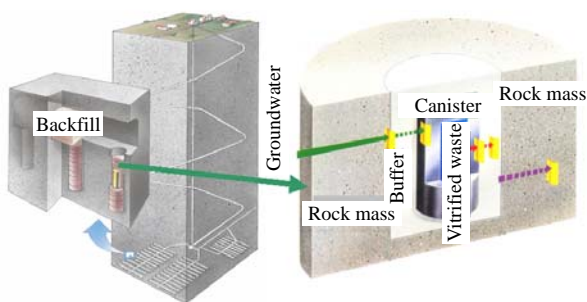


Fig.1 Conceptual model for China's high-level radioactive waste disposal

The buffer is one of the main engineered barriers for HLW repository. It was early concluded that clay-based embedment of canisters and backfill in drifts and shafts offers the best isolation through the low hydraulic conductivity and canister supporting ability of such materials. Assessment of the various properties had led to the conclusion that montmorillonite-rich clays are most suitable for preparing the buffer material. Bentonite would appear to fulfill the criteria placed on the buffer material in a deep repository for high-level nuclear waste^[2-4]. The buffer material is expected to maintain its low water permeability, thermal conductivity, self-sealing, radionuclide sorption and retardation, chemical buffering, canister support and stress buffering properties over a long period of time^[5].

The scientific scope of buffer/backfill materials study is summarized by the author, as shown in Fig.2.

In China, from 1985 on, Chinese scientists began to study the bentonite's behavior and its feasibility as buffer material in HLW repository. The related scientific activities cover literature investigation and use of inorganic sorbents as backfill materials for underground repository^[6], bentonite deposit screening^[7], study of basic property of GMZ Ca-bentonite^[8] and GMZ Na-bentonite^[9] and comparison study of basic property between GMZ - 1 and KunigelV1 bentonite^[10]. GMZ deposit is selected as a candidate buffer supplier in China in 1996^[7] and GMZ - 1 Na bentonite was selected as a reference material for Chinese buffer/backfill study in 2002^[9].

2 GMZ - 1 BENTONITE

GMZ - 1 bentonite is Na-bentonite produced from GMZ deposit and selected as reference material for Chinese buffer material study.

The mineralogical composition of GMZ - 1 has been quantitatively analyzed by X-ray diffraction

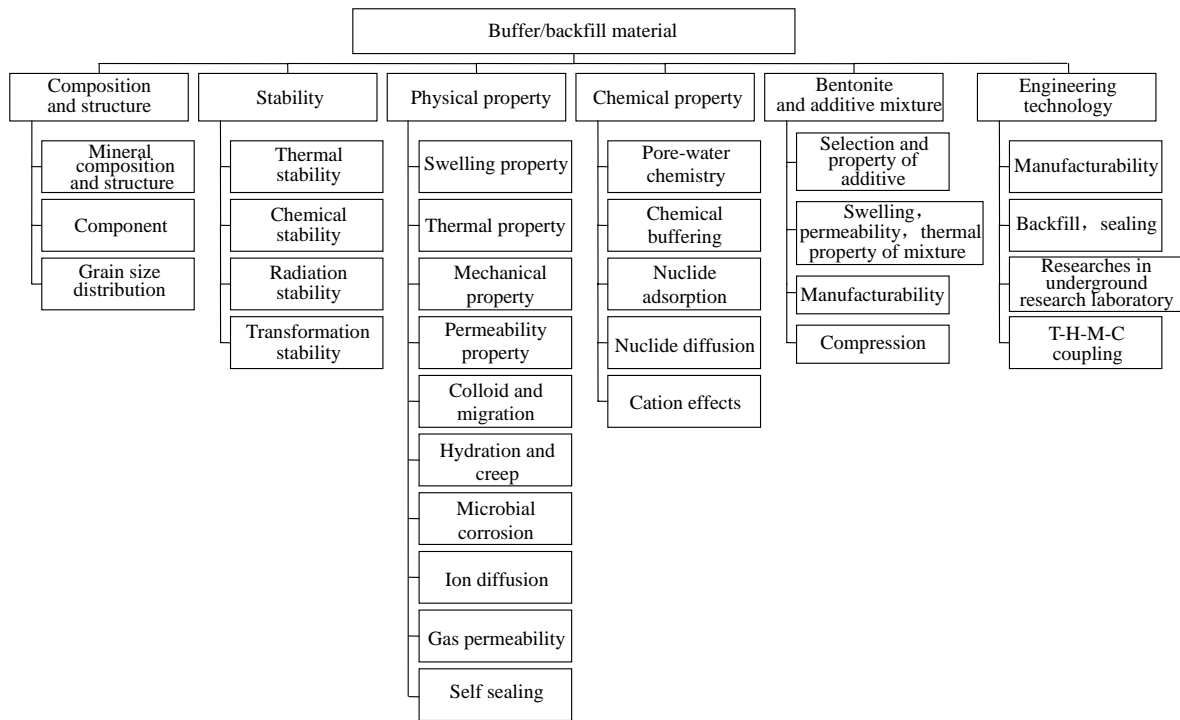


Fig.2 Scientific scope of buffer/backfill materials study

analyses. The bulk composition was determined as: quartz 11.7%, cristobalite 7.3%, feldspar 4.3%, calcite 0.5%, kaolinite 0.8%, and montmorillonite 75.4%.

The basic feature of GMZ - 1, the reference material for Chinese buffer/backfill material is summarized in Table 1.

Table 1 Basic parameters of GMZ - 1 bentonite

Montmorillonite content/%	Methylene blue exchange capacity / $(\text{mmol} \cdot (100 \text{ g})^{-1})$	Cation exchange capacity / $(\text{mmol} \cdot (100 \text{ g})^{-1})$	Real density / $(\text{g} \cdot \text{cm}^{-3})$	Alkali index
75.4	102	77.30	2.66	1.14
pH	Exchangeable cation/ $(\text{mmol} \cdot (100 \text{ g})^{-1})$			
	$E(\text{k}^+)$	$E(\text{Na}^+)$	$E(1/2\text{Ca}^{2+})$	$E(1/2\text{Mg}^{2+})$
8.68 - 9.86	2.51	43.36	29.14	12.33

3 PHYSICAL PROPERTIES OF GMZ - 1

The bentonite buffer material around the HLW canister has a key role for repository safety. It must hold the canister in the center of the disposal pit. The buffer will also have to conduct the fuels' residual heat. The bentonite will hinder groundwater, possibly containing various corrosive substances from flowing

freely to the canisters' surface. Another key property of the buffer material is its ability to swell by water uptake; therefore it fills voids in the engineered barriers and fractures in the surrounding host rock.

Thus, in case of Chinese candidate buffer material: GMZ - 1, basic parameters related to swelling, thermal, hydraulic and mechanic properties should be obtained in order to assess its feasibility to be served as buffer in HLW repository.

3.1 Thermal properties

The buffer is required to have a good thermal conductivity in order to promote effective heat transfer to the surrounding rock. This keeps temperatures in the waste package low, preventing recrystallization of glass that might be caused by elevated temperatures arising from the radiogenic heat of the vitrified waste. As the buffer is not completely saturated immediately after emplacement of the waste packages, the thermal properties of unsaturated buffer should be specified in the design^[11]. Therefore, thermal conductivity as function of dry density(1.4, 1.6, 1.8 g/cm³) and water content(9%, 14%, 18%, 24%) was measured by surface heat source method in this study. The instrument is thermophysical Properties Analyzer TPA 501(Fig.3). The experimental results are given in Table 2.

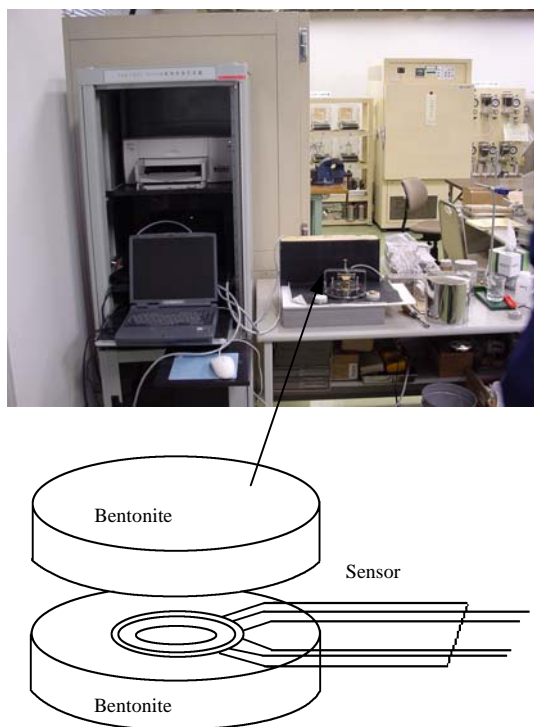


Fig.3 Instrument for thermal conductivity test

Table 2 Results of GMZ - 1 thermal conductivity tests with dry density and water content

Dry density/(g · cm ⁻³)	Water content /%	Thermal conductivity /(W · (m · K) ⁻¹)
1.4	8.76	0.57
	13.83	0.83
	18.17	0.97
	24.07	1.19
1.6	8.64	0.80
	13.56	1.10
	17.94	1.30
	26.70	1.51
1.8	8.61	1.02
	13.36	1.43

The thermal properties of the buffer depend on factors such as density, void ratio and water content, in a similar manner to general soil materials^[5]. The thermal conductivity of GMZ - 1 increases as the dry density increases for the same water content; and the thermal conductivity increases as the water content increases for the same dry density.

3.2 Hydraulic properties

When the waste package is emplaced, the buffer is in an unsaturated condition suitable for transportation and handling. After that, groundwater is drawn into the buffer by capillary action, the voids become filled with groundwater and the buffer becomes saturated. Therefore, it is necessary to understand the hydraulic properties for both the saturated and unsaturated state. In this study, The experiments on the density-dependence and the temperature-dependence of the saturated hydraulic conductivity of GMZ - 1 have been conducted.

Permeability tests are conducted according to the Japanese Geotechnical Society Standard(JGS T311). The test cell of diameter 50 mm and depth 10 mm is filled with test material at specified density(1.4, 1.6, 1.8 g/cm³) and set in a thermostatically controlled heating element. At each temperature in the stepwise heating process(25 °C → 60 °C → 90 °C), distilled water is injected through the test sample by compressed air of 0.3 MPa(1.4 g/cm³) /0.6 MPa(1.6, 1.8 g/cm³) and the outflow rate is measured with an electronic balance(Fig.4).

The saturated hydraulic conductivity $k(m \cdot s^{-1})$ is calculated from the outflow rate $Q(m^3 \cdot s^{-1})$ by the following equation in accordance with Darcy's law:

$$Q = kiA \tag{1}$$

where A is cross-section area of the test sample(m²), and i is hydraulic gradient.

Experimental results are shown in Table 3. The hydraulic conductivities of GMZ - 1 increase as the temperature increases, and decrease as the dry density increases.

3.3 Mechanical properties

The buffer material should provide sufficient mechanical supports of the overpack to ensure the stability^[12]. Mechanical property includes compressive strength, modulus of elasticity, shear properties, consolidation properties and tensile strength, etc.

Unconfined compression strengths of GMZ - 1 as function of dry density(1.4, 1.6, 1.8 g/cm³) and water

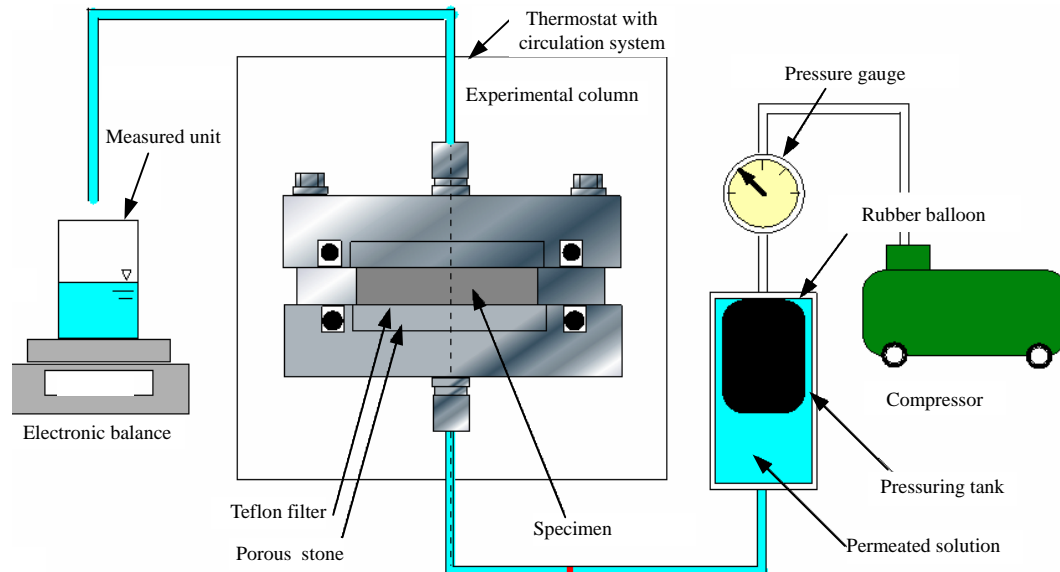


Fig.4 Schematic view of equipment for hydraulic conductivity test

Table 3 Results of GMZ - 1 hydraulic conductivity tests with dry densities and temperatures

Dry density /($\text{g} \cdot \text{cm}^{-3}$)	Temperature / $^{\circ}\text{C}$	Hydraulic conductivity /($\text{m} \cdot \text{s}^{-1}$)
1.4	25	1.12×10^{-12}
	60	1.87×10^{-12}
	90	2.91×10^{-12}
1.6	25	1.94×10^{-13}
	60	3.61×10^{-13}
	90	5.75×10^{-13}
1.8	25	9.99×10^{-14}
	60	1.99×10^{-13}
	90	3.00×10^{-13}

Table 4 Results of GMZ - 1 unconfined compression tests with dry densities and water contents

Real dry density /($\text{g} \cdot \text{cm}^{-3}$)	Water content /%	Unconfined compression strength /MPa
1.4	8.32	0.96
	13.24	1.37
	17.45	1.05
	22.80	1.04
1.6	8.29	3.06
	13.06	3.43
	17.43	2.37
	23.58	1.74
1.8	8.51	6.50
	12.80	7.40
	17.09	5.15

content(9%, 14%, 18%, 24%) at room temperature was conducted in this study. The instrument is AG - 10TB. The cylindrical specimen size is 30 mm (diameter)×60 mm(height). The strain rate is approximately to be 1×10^{-2} /min. The test method and procedures basically conform to the Japanese Geotechnical Society Standard(JGS T511). Data are given in Table 4.

In general, with the same water content, a larger initial dry density leads to a larger unconfined compressive strength, and with the same dry density,

a larger water content leads to a less unconfined compressive strength. The unconfined strength for GMZ - 1 with 12.8% - 13.24% water content is higher than that with other water content for the same dry density, which might be related to the optimum water content.

3.4 Swelling characteristics

A key property of the buffer material is its ability to swell by water uptake, thereby filling voids in the

engineered barriers and fractures in the surrounding host rock.

Thus, parameters related to swelling property are necessary to the design of buffer material. Since the stress applied on the confined boundary surface when a bentonite specimen is infiltrated with water is not always equal to the swelling pressure^[11] and so the applied stress on this confined boundary surface is defined here as the “swelling stress”^[12]. Swelling stresses as function of dry density(1.2, 1.4, 1.6, 1.8 g/cm³) were measured in this study. The instrument for swelling stress measurement is shown in Fig.5; and the corresponding data are given in Table 5.

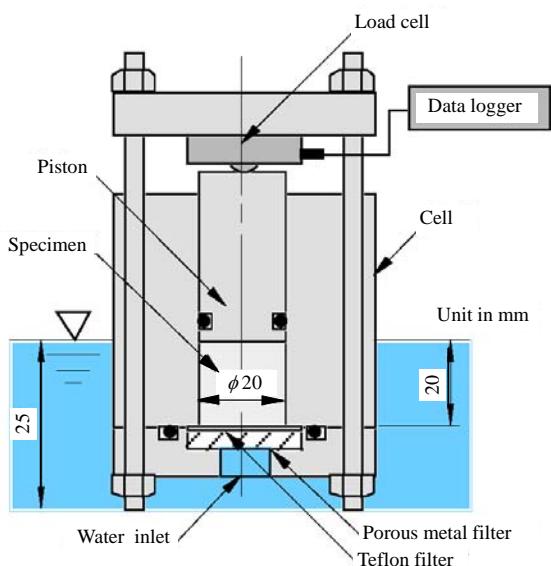


Fig.5 Test apparatus for measurement of swelling stress^[12]

Table 5 Swelling stresses as a function of dry densities

Dry density/(g • cm ⁻³)	Swelling stress/MPa
1.2	0.09
1.4	0.58
1.6	3.17
1.8	10.10

Table 5 shows that the swelling stress increases as dry density increases.

4 CONCLUSIONS

The deep geological disposal is regarded as the most reasonable and effective way to safely disposal

high-level radioactive wastes in the world. The conceptual model of geological disposal in China is based on a multi-barrier system and buffer/backfill material is one of the most important engineering barriers in the HLW repositories. Due to the very low permeability and excellent retardation of nuclides from migration etc., the bentonite is selected as base material of the buffer/backfill material in HLW repositories. GMZ deposit is selected as the candidate supplier for buffer material of HLW repositories in China. From 2000 on, systematic study has being conducted on GMZ - 1 that is Na-bentonite produced from GMZ deposit and selected as reference material for Chinese buffer material study. GMZ - 1 bentonite is characterized by high content of montmorillonite(about 75%) and less impurities. The systematic study on GMZ - 1 is essential to safe dispose the high-level radioactive wastes in China.

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