Deep Geological Disposal

Swelling Clay based Buffer Component of Engineered Barrier System of Waste Disposal Facilities

Prateek Srivastava[1], Shiva Kumar[1], T. K. Pal[1], R. K. Bajpai[1,3]* and C. P. Kaushik[2,3]

[1]Technology Development Division, Bhabha Atomic Research Centre, Mumbai-400085, INDIA
[2]Waste Management Division, Bhabha Atomic Research Centre, Mumbai-400085, INDIA
[3]*Homi Bhabha National Institute, Mumbai-400094, INDIA

Barmer clays of Akli formation, Rajasthan

ABSTRACT

Swelling clays in general and Na-Smectite clays in particular have emerged as highly promising candidate material for use as buffer component of Engineered Barrier System (EBS) in deep as well as shallow radioactive waste disposal facilities due to their very low permeability, high retention of radionuclides, geochemical compatibility with host environment and high plasticity required for providing cushioning to disposed waste packages against rock movements. Such clays are under extensive characterization worldwide for assessment of their suitability for use as buffer material in waste disposal facilities. In the Indian context, a Deep Geological Disposal Facility (DGDF) with a capacity of accommodating 10000 waste canisters would require about 0.1 million tonnes of good quality swelling clays with a specific range of thermal-mechanical-hydraulic-radiological properties. Extensive experimental and numerical simulation based studies have been performed on promising Na-Smectite clays of Barmer district, Rajasthan to establish their suitability for the above purpose. The studies reveal that these clays possess adequate chemical, thermal and radiological properties on par with international reference clays MX-80 of the United States of America to serve as buffer material in Indian radioactive waste disposal facilities. It is also established that 25cm thick layer of Na-Smectite clay with high Smectite content (~80%) compacted to a dry density of 1.6gm/cc with inter-canister spacing of 2m can provide a stable thermal field around disposed waste canister by maintaining temperatures well below 100°C at any point of time with minimum possibility of building of thermal stresses and resultant micro-fracturing in EBS as well as in host rocks.

KEYWORDS: *Smectite, Geological disposal, Engineered Barrier System, Vitrified radioactive waste, Akli formation.*

Introduction

Disposal of solid radioactive wastes essentially requires their long term confinement and isolation from the accessible environment over varying extent of time span ranging from few hundreds of years to millions of years depending on half lives and concentrations of key radionuclide contained within such wastes. These objectives are achieved by disposing these wastes at varying depths underground. Low level solid wastes with short lived radionuclides like Cs-137, Sr-90 are thus disposed in engineered structures built within depth ranges of few tens of meters whereas vitrified high level waste with long lived radionuclides like isotopes of actinides, activation and fission products *viz.*, americium, neptunium, uranium, nickel, molybdenum, iodine, technetium, tin etc., become target wastes for disposal at >500m depth in specifically designed and excavated underground structures within suitable host rocks popularly known as Deep Geological Disposal Facilities (DGDF).

Sites for building these underground disposal facilities are selected based on very extensive evaluation of various parameters and earth processes that would eventually control the confinement of wastes over the desired period of time. Depth, geological stability of the site, low permeability & high strength of host rocks, high sorption capacity for radionuclides etc. are among important parameters that ensure long term isolation of wastes[1]. The long term safety offered by these disposal facilities are invariably provided by a combination of geological barrier (host rock) and Engineered Barrier System (EBS) represented by waste form, canister, over pack and additional layers of protection between the waste packages and the site soils/rocks in the form of admixtures of natural

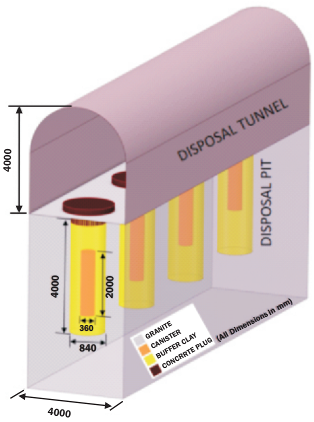

Fig.1: Layout of disposal concept adopted in India.

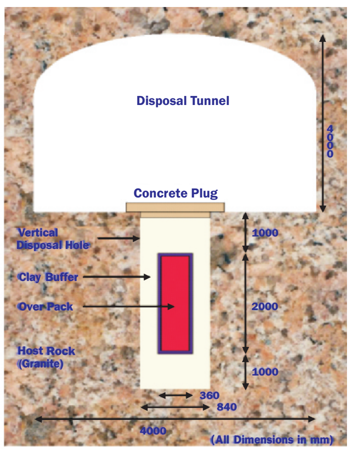

Fig.2: Schematic view of vertical disposal pit.

*Author for correspondence: R. K. Bajpai
E-mail address: rkbajpai@barc.gov.in

Fig.3: Barmer clays of Akli (a village) formation in Rajasthan.

Table 1: Some important temperature-dependant geo-technical properties of Barmer clays

Sl. No.	Temp. (°C)	Specific Gravity (gm/cm^3)	Liquid limit (%)	Plastic limit (%)	Optimum moisture content (%)	Dry density (gm/cm^3)	Specific surface area (m^2/gm)	Free Swell index (%)
1	65	2.785	328.37	35.29	36.18	1.182	448	776
2	100	2.778	305.97	34.26	36.07	1.276	394	753
3	125	2.765	292.62	33.09	35.2	1.316	371	734
4	160	2.762	291.66	32.73	34.84	1.317	353	723

clays and sand, crushed rocks, crushed rocks and clay admixtures. These layers are referred as buffer material. A reference disposal system under consideration in India is shown in Fig.1 & Fig.2 [2]. Choice of the natural material is controlled by the site geochemical environment and groundwater conditions. In India, extensive works on clay-sand based EBS have been taken up in two decades[3, 4]. Studies have revealed that granite host rock based Indian Deep Geological Disposal Facility with a capacity of holding 10000 waste loaded canisters would require 0.1 million tonnes of high quality clays for use as buffer and backfill[5].

Waste loaded canisters at the time of their disposal in DGDF are characterised by continuous heat flux of the order of 500 W mainly produced by radioactive decay of fission products contained in the waste. As Clay based EBS lies in direct contact of the canister, their important functions include smooth dissipation of the heat from the waste into the surrounding rock, arrest the ingress of groundwater towards the waste package, protect the waste canister against rock movement, and retard the transport of radionuclide that may eventually release from the waste canister in the distant future. In India, scheme of deep disposal involves emplacement of vitrified high level radioactive waste contained in SS Canisters housed in 2m long metallic over pack at depth of 500 to 700m in a disposal pit of 4m depth and 840mm diameter with layers of swelling clay based EBS [2]. The Na-Smectite clay layers in a DGDF are expected to withstand about 15 MPa combined thermal and mechanical stresses coupled with a temperature of the order of 90°C [6].

In addition to these, they are also expected to witness varying geochemical condition like pH, Eh, changing groundwater compositions, oxygen fugacity etc.

Development of Clay Buffers of EBS

Large deposits of swelling clays chiefly composed of Na-Smectite, from the Barmer district of Rajasthan, known popularly as Bentonite, have been taken up for detailed evaluation. Samples of swelling clays used in this study were collected from the Akli mine located in the Akli village, Barmer district, Rajasthan, India. The Na-smectite rich clays in this area belong to the Akli Formation of Jagmal Group and are of Palaeocene age. The sampling was restricted to clay horizons with visible homogeneity in terms of texture, grain size, colour etc to avoid mineralogical variations that may result in wide scatter in parametric values. The samples were tested for their thermal, mechanical and hydraulic parameters following standard testing protocols mainly ASTM, ISRM and IS. Important characteristics of these clays are given in Table.1.

Limited decrease in moisture content (3.70%) and swelling index (6.82%) with increasing temperature make these clays highly suitable for use as buffer material in DGDF. Similarly, decrease in specific surface area and increase in dry density are also within desired limits. The UCS of these compacted clays varies from 2 to 3 MPa. The swelling pressure of these clays range from 2100-2200 kPa with a saturated hydraulic conductivity 1-5x10^{-15}cm/s. These parameters compare well with MX-80 clays of Wyoming Basin USA, which have emerged as the most suitable clays for use in DGDF[7, 8].

Mineralogy and Geochemistry

Chemical composition of these clays is characterized by 48-45% SiO_2, 15-20% Al_2O_3 and Fe_2O_3 3-6% with 3-4% MgO and 1-2% Na_2O and almost 4% organic impurities. XRD analysis of samples confirms presence of Na-Smectite as the major mineral (60-85%) accounting for higher SiO_2 and Al_2O_3. The Smectite content of these clays is adequate and at par with

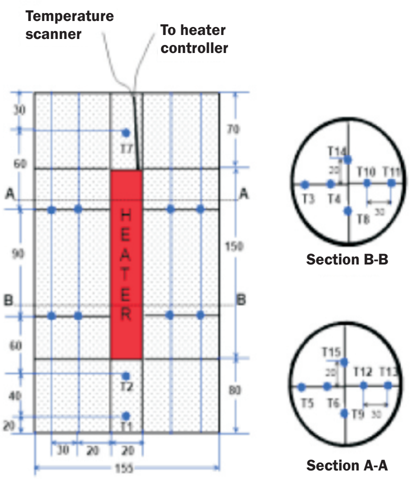

Fig.4: Layout of the TMH Experiment.

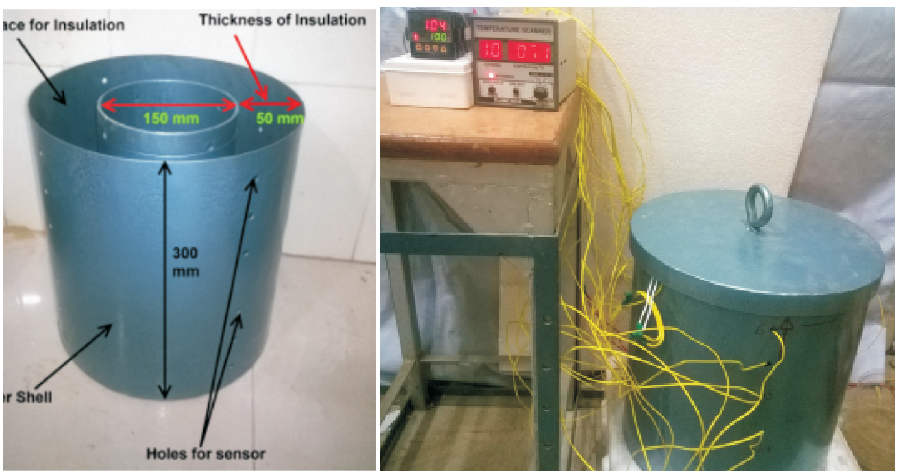

Fig.5: Components of Temperature-Moisture-Hydraulic (TMH) experiment.

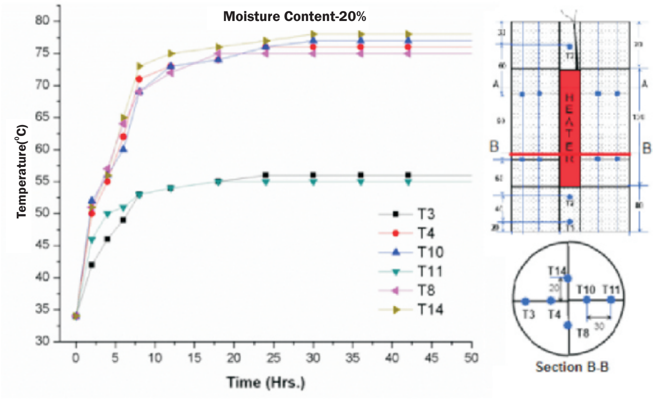

Fig.6: Radial temperature along section B-B within compacted buffer at moisture content 20% .

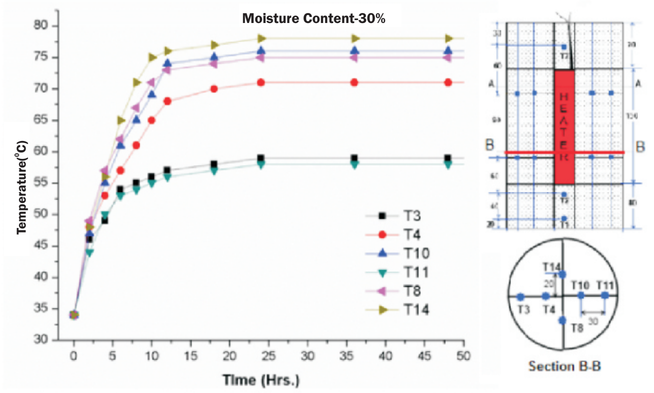

Fig.7: Radial temperature along section B-B within compacted buffer at moisture content of 30%.

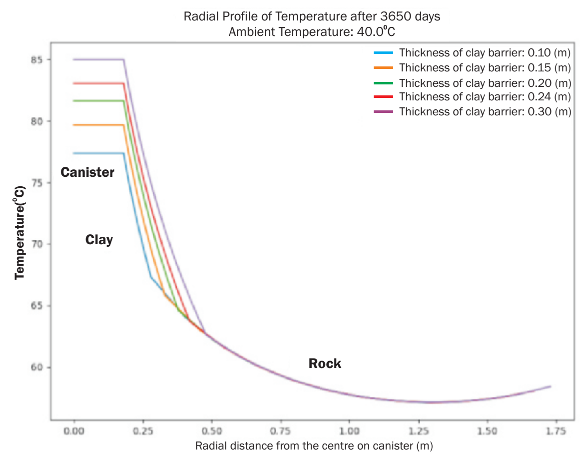

Fig.8: Radial profile of temperature at 5 different spacings of two disposal pits.

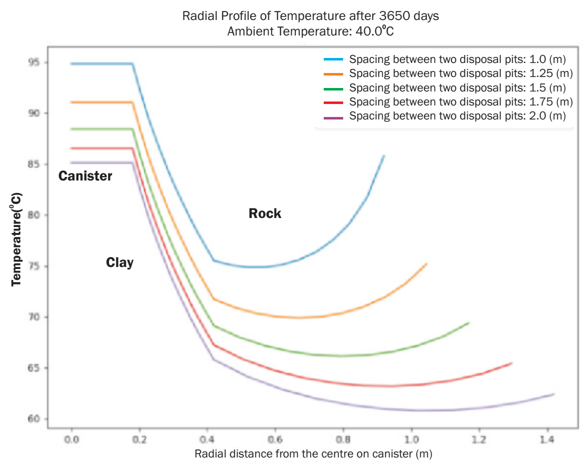

Fig.9: Radial profile of temperature at 5 different thicknesses of clay barrier.

Table 2: Thermal and physical properties of host rock (granite) & Barmer clay

Properties	Host rock (granite)	Barmer clay
Thermal conductivity (W/m/K)	3.3	1.27
Specific heat (J/Kg/K)	1000.0	870.0
Density(Kg/m^3)	2580.0	1600.0

MX-80. Temperature dependant variation in Cation Exchage Capacity (CEC) values reveals initial increase in the CEC from 54 to 59.52 up to temperature of 60°C which reduces to 55.36 at a temperature of 150°C. Experimental studies on sorption of americium on these clays have also been taken up. At lower pH values (2.5-3.5), the sorption of Am(III) increases slowly but rises sharply between the pH ranges of 4–8. Beyond a pH >8 the sorption of Am(III) remains nearly constant. The Kd value for Am(III) range of 111360-612023 mL/g at lower pH (2.5-4.0), 156828-918997 mL/g in intermediate pH range (4-8) and almost constant Kd (918997-926447 mL/g) at pH>8. These results indicate stability of clays in hyper alkaline environment that may eventually develop in disposal facilities due to the presence of concrete and cementeous material. In addition to this, this clay remains unaffected in the presence of anions like chlorine, sulphate and nitrate, typical of granite waters at higher pH values[9].

Experimental Evaluation

For experimental evaluation of the heat distribution within the clay layers of EBS, an experimental set up was developed. It includes a carbon steel cell of 300 mm length and 155mm diameter which was filled with Na-Smectite compacted to a dry density of 1.6 gm/cc with water saturation of 8.5%. The evolution of temperature in compacted bentonite depends on the dry density of clay and moisture content. A 150mm long and 20mm diameter heater with 500 watt output was used as heat source replicating the high level radioactive wastes. A total of 13 thermo couples were installed in the cell at locations shown in Figs. 4 & 5. The experimental dimension, heating system and hydraulic properties of clays were optimised through extensive sample scale testing as well as numerical simulation to optimise various components so that a reasonable replication of actual geological system is accomplished at experimental scale.

The heater remained on for 432 hours at a designated temperature and thereafter it was switched off after attaining steady state. Typical experimental results of temperature measurements with buffer zone measured for initial saturations are shown in the Figs. 6 & 7. The temperature evolution is highly influenced by the presence of moisture on the Na-Smectite clays and can be clearly observed from temperature profile obtained at different monitoring points for moisture content of 20% and 30% respectively. In case of 30% moisture, the temperature curves show larger spacing as compared to those for clays with 20% moisture. Larger movement of moisture is recorded in high temperature zone which also enhances the heat transfer and causes an increase in temperature. Slow evolution of temperature is due to the small temperature gradient. At all the locations within the clay buffer, the initial rate of heat transfer was comparatively high mainly due to the presence of moisture. This is mainly accomplished by evaporation near the heater contact and distribution of moisture in distant areas. Experiment reveals that temperature within the clay remains below 100°C under varying moisture content and hence establishing the capability in smoothly conducting the heat through them.

Modelling

One of the important parameter controlling suitability of these clays for their use as buffer material around disposed waste canisters is their heat dissipation capacity. Poor heat dissipation through clay buffer results in heat build up around disposed waste and may eventually lead to micro-cracking of barrier its self as well as surrounding host rock.

To model heat dissipation through a 25 cm radial thickness of buffer (Barmer Na-smectite) clay layer, a case involving three waste loaded canisters of 2m length and 30cm diameter with varying inter-canister spacing and buffer thickness was analysed. The material properties used in the modelling are shown in Table 2. The conductive heat transfer process is mathematically modeled using Fourier's Law. The governing heat diffusion equation is first solved for an instantaneous finite height vertical line heat source representing the actual canister using Laplace transformation technique. Since the governing equation of the heat transfer for a single canister is a linear partial differential equation, the temperature field for multi-canisters disposal in DGDF is calculated by superposing the solution of a number of single line heat sources.

A computer program is written for the closed form solution of temperature. The integration part of the solution is carried out numerically using the Gaussian quadrature integration technique. The graphical user interface (GUI) is developed using the Tkinter module of Python. The simulation was run for 5 different spacing between two disposal pits as 1.0m, 1.25m, 1.5m, 1.75m, 2.0m and 5 different values of radial thickness of Na-Smectite clay barrier as 0.1m, 0.15m, 0.2m, 0.24m, and 3.0m as shown in Figs. 8 & 9. The analysis reveals that a spacing of 1.0m between two disposal pits results in interaction of their thermal field with resultant temperature rising above 100°C. However, for spacing of >1m, Barmer Na-Smectite clay buffer provided adequate heat dissipation to maintain temperatures within recommended limit of 100°C. The variation of thickness of clay barrier does not have impact on the temperature of the rock but it increases the canister surface temperature.

Conclusions

Extensive laboratory based sample testing of swelling Na-Smectite clays of Barmer, Rajasthan for their TMH parameters, their performance under heat-hydraulic load in meter scale experiment and field scale numerical simulations of heat field around disposed waste canisters surrounded by clay based EBS has demonstrated suitability of these clays for use in Deep Geological Disposal Facility for ultimate disposal of heat emitting vitrified high level waste. These clays with almost 80% Smectite and adequate physicochemical and geotechnical parameters are comparable with those of MX-80 clays from the Wyoming Basin USA, an internationally recognised buffer clay. The studies establish the adequacy of 25cm radial thickness of these clay buffers and 2m inter-canister spacing for ensuring temperature limits within 100°C in any part of the waste disposal facility.

Acknowledgements

Authors acknowledge the valuable contributions made by a number of researchers from Indian Institute of Science, Bangalore, Indian School of Mines, Dhanbad, IIT Patna and IIT Bombay in these studies. Authors express their sincere thanks to RACD, BARC for contributions in radiological characterization. Rajasthan Mineral Development and AMDER Hyderabad have provided support during the field investigations for which authors are indebted to them.

References

[1] R. K. Bajpai, P. Shrivastava, P. K. Narayan, K. Banerjee "Host Rock Characterization, In-Situ Experiments, Numerical TMH Simulations and Natural Analogue Studies in the Indian Deep Geological Disposal Program" in Boris Faybishenko, Jens Birkholzer, David Sassani, Peter Swift (eds.) Fifth World Wide Review of Geological Challenges in Radioactive Waste Isolation, LBNL-1006984, California, 2016, 11.1-11.9.

[2] P. K. Narayan, Bajpai R. K. Deep Geological Repositories for Vitrified High Level Long Lived Wastes. *IANCAS Bulletin,* **VI (3)**, 2007, 224-237.

[3] Sudhakar M. Rao, K. Ravi "Hydro-mechanical characterization of Barmer 1 bentonite from Rajasthan, India". *Nuclear Engineering and Design.*, 2013, **265**, 330–340.

[4] Sharau Kesar, Sumeet Kumar, Bajpai R.K., Tomar B.S. "Diffusion of Na(I), Cs(I),Sr(II) and Eu(III) in smectite rich natural clays". *Jour. of Environmental Radioactivity* (Springer), 2015, **151**, 216-223.

[5] R. K. Bajpai "Progress in implementing Deep Geological Repository in India". International Atomic Energy Agency Technical Meeting on Global Progress on Geological Disposal of Radioactive Waste, GCNEP New Delhi, 22-26 May 2018.

[6] S. Maheshwar, A. K. Verma, T. N. Singh & R. K. Bajpai "Study of thermo-hudro-mechanical processes in conceptual Indian nuclear waste repository". *J. Earth Syst. Sci*, Indian Academy of Science, 2015, **124**(8), 1693-1708.

[7] Perdrial N. Julia, Warr N. Laurence, "Hydration Behavior of MX80 Bentonite in a Confined-Volume System: Implications for Backfill Design". *Clays and Clay Minerals*, 2011, **59**(6), 640–653.

[8] L. Xiaodong, R. Prikryl, R. Pusch "THMC testing of three expandable clays of potential use in HLW repositories". *Applied Clay Science*, 2011, **52**, 419-427.

[9] S. Kumar, A. S. Pente, R. K. Bajpai, C. P. Kaushik, B. S. Tomar, "Americium sorption on smectite-rich natural clay from granitic ground water" *Applied Geochemistry*, 2013, **35**, 28–34.