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Temperature effects on the erosion of bentonite

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ABSTRACT

Compacted bentonite is a candidate barrier material (buffer, backfill and plug), a critical component of engineered barrier systems for geological disposal of high-level (heat generating) radioactive waste. The clay barrier can be subject to erosion due to interactions with groundwater at the interface between the clay and fractured host rock. The engineered barrier system undergoes a thermal period (~5k-10k years) at which potential mass loss and erosion assisted piping of clay buffer can happen under elevated temperature. This paper presents the results of laboratory experiments on the erosion of compacted bentonite at ambient and elevated temperatures. Compacted samples of Volclay bentonite (dry density 1.45 Mg/m^3) were subject to a constant flow of deionised water fat a porous boundary for three months. Erosion and swelling pressure were simultaneously recorded using a bespoke constant rate of strain system (CRS) by which the eroding conditions were created at the bottom of clay sample. The results show that the mass loss increased by approximately three times at the elevated temperature (at steady state). The outflow fluid was collected at regular intervals and the eroded materials were characterised by Dynamic Light Scattering to develop an understanding of the sizes and concentrations of particles and their evolution. It was found that the particle sizes in colloidal solution range between 237 nm and 1060 nm whilst the average size was not considerably affected by the elevated temperature. The results presented provides new insights into the erosion phenomena at elevated isothermal temperature in smectite rich clays.

Keywords: geological disposal, compacted bentonite, erosion, elevated temperature, mass loss

1 INTRODUCTION

Geological disposal concepts are internationally recognised as the preferred solution for long-term management of the high-level (or higher activity) nuclear waste (HLW). In geo-disposal concepts, the release of radionuclides from the waste to biosphere is minimised by a multiple layers of barriers. Smectite-rich clays (bentonite) are considered as candidate material for the buffer/backfill and plug in this concept; especially in repositories constructed in fractured/crystalline rock. Swelling clays play important functions in the engineered barrier system as the buffer, backfill and also the sealing/plug material. However, the erosion of clay buffer is likely to occur due to hydro-chemical interactions of the clay barrier with groundwater at the interface between with the host rock. Erosion can create potential pathways for radionuclides to migrate from the waste form into the surrounding rock which has potentially significant environmental and public health impacts. Therefore, developing prediction capability for the clay buffer erosion and associated transport phenomena is critical in order to develop the safety case and performance assessment of the geological disposal.

The clay barrier can experience a long period at elevated temperature environment (sourcing from the hot canister) at which the re-saturation happens. Erosion at re-saturation stage can initiate and progress when temperature at the interface between buffer and rock is at elevated level. During the thermal period that may take 5000-10,000 years, temperature of the buffer at the interface with rock can increase (e.g.

up to 60°C and depending on the concept). Notably, high temperatures might also be expected for the clay barrier in other applications in geological disposal such as backfill or plug. The mass loss of the clay barrier by erosion can be initiated and extended during the thermal period. A large body of experimental research has developed in the last 20 years or so that provides critical knowledge about the erosion of bentonite barriers in the context of geological disposal. Various aspects of erosion and influential parameters (e.g. water flow rate, compaction level, the effects accessory minerals) have been studied but almost all studies focus on the erosion at ambient temperature. The aim of the research presented in this paper has been to develop preliminary observations and fundamental insights into the overall impacts of temperature on erosion of compacted bentonite and the characteristics of the eroded particles.

The experimental studies in this field have significantly improved the understanding of the erosion of compacted bentonite and influential parameters such as flow rate, salinity or ionic strengths and the boundary/geometrical conditions (e.g. Pusch, 1999; Sane et al., 2013; Moreno et al., 2011; Asensio et al., 2018). This understanding has also been translated into limited predictive models for erosion of clays (e.g. Yan et al., 2021; Sedighi et al, 2022). It is known that high temperature affects the behaviour of compacted bentonite in a complex and highly non-linear manner (e.g. Sedighi et al., 2018) and experimental observations provide evidences that elevated temperature can influence the swelling and extrusion, but the knowledge about its potential impact on erosion is very limited.

Figure 1 provides an illustration of the erosion of bentonite buffer and conceptualises the three main physical processes controlling the overall erosion of swelling clay: (i) free swelling and extrusion of the clay to the fractures, representing a continuous transformation to clay gel; (ii) detachment of clay particles, representing the formation of clay sol; and (iii) migration of detached particles with the water flow (Sedighi et al., 2022). For the individual processes presented in Figure 1, high temperature can be influential. Observations of the impacts of temperature on swelling pressure development reported in literature provide some indirect insights into the effects of extrusion behaviour at high temperature but there is still a lack of understanding of the overall impacts of elevated temperature on erosion; especially during non-isothermal re-saturation phase of the buffer. At this operational stage; potential mass loss of the clay barrier and erosion assisted piping during re-saturation are important and less understood compared to other coupled hydro-mechanical processes in compacted bentonite.

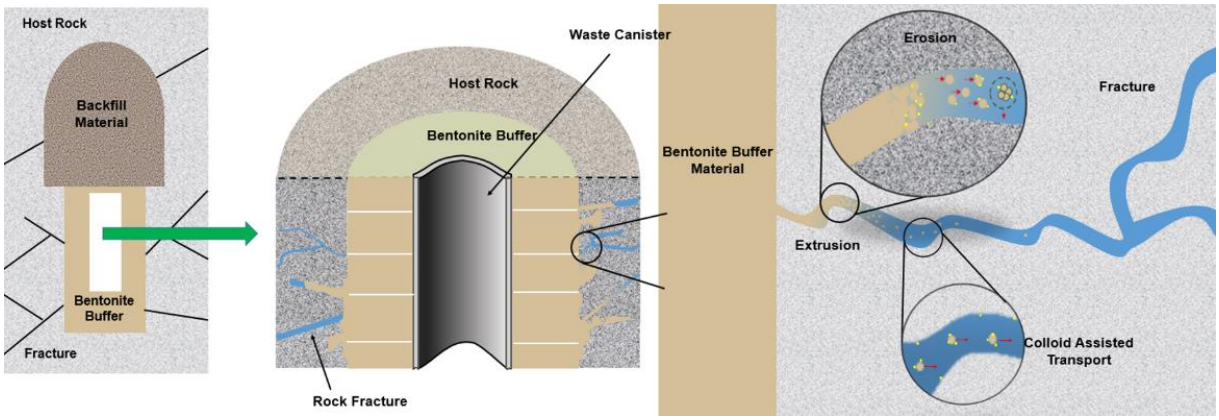


Figure 1. *Schematic of the erosion of compacted bentonite (example of a buffer) at the interface with fracture rock. The main mechanisms involved are shown in the right image.*

This paper presents the results of a series of erosion tests on compacted bentonite at ambient temperature (25°C) and elevated temperature (60°C). Compacted samples of Volclay bentonite (dry density 1.45 Mg/m^3) were subject to a constant flow of deionised water through a porous boundary for three months. Erosion and swelling pressure were simultaneously recorded using a bespoke constant rate of strain system (CRS) by which eroding conditions were created at the bottom of clay disk. Erosion and swelling pressure development have simultaneously been monitored for nearly 3 months and the eroded particles have been analysed at certain time intervals.

2 MATERAIL AND METHODS

2.1. Clay properties

The clay used in this study was powdered bentonite (Volclay SPV200) supplied by AMCOL (UK). Its properties are summarised in Table 1. The bentonite was a mixture of dioctahedral montmorillonite with small amounts of quartz, plagioclase, K-feldspar, pyrite, gypsum, opal-C/cristobalite and traces of kaolinite. The clay fraction less than 2 μm of bentonite is a mixture of montmorillonite with traces of illite/mica and kaolinite. The analysis of the chemical compositions of bentonite powder was carried out using Energy-dispersive X-ray fluorescence (EDXRF). The hygroscopic water content was determined by oven drying at 105°C. The particle size distribution was obtained by dry sieving and hydrometer tests. Liquid and plastic limits, and the specific gravity of the soil grains were obtained following relevant British Standards. The measurement of the total surface area was determined by the adsorption of ethylene glycol monoethyl ether (EGME). The cation exchange capacity and exchangeable cation composition of the clay were obtained by displacing the exchangeable cations by 1 M ammonium acetate (NH_4OAc) at pH=7.0 and analysed by inductively coupled plasma optical emission spectroscopy (ICP-OES).

Table 1. *The physical and chemical properties of the bentonite clay used in this study.*

Properties	Quantity
Clay fraction (%)	56
Montmorillonite content (%)	86.8
Accessory minerals (%)	
Quartz	3.9
Plgogioclase	5.6
K-feldespar	0.9
Pyrite	0.2
Gypsum	Trace
Hygroscopic water content (%)	12
Liquid limit (%)	409
Plastic limit (%)	52
Specific gravity (G_s)	2.74
External surface area (m^2/g)	20.0
Total surface area(m^2/g)	528
CEC (meq/100g)	94
Exchangeable cations (meq/100g)	Na^+= 56, Ca^{2+}= 30, Mg^{2+}= 7, K^+=1.0

2.2. Erosion setup and testing method

A bespoke high pressure, high temperature constant rate of strain system (CRS) was refurbished to accommodate the possibility of experimenting the erosion. The colloid generation was studied by the system in which the conditions for erosion were created at the bottom of the clay disks while the swelling pressure development was monitored at the top of the samples. Figure 2 presents a schematic diagram of the experimental setup designed of this study.

To create the eroding condition at the bottom of the sample, sintered stainless-steel filter was placed at bottom which follows a similar porous system used by Missana et al. (2011). The average or nominal pore size of this plate was 100 μm. The two top openings were closed, and a filter paper and a thin film were placed on the top of the specimen to ensure there was no outflow through the top of the sample. Bentonite specimens were formed by compacting statically the powder to the target dry densities using a specimen ring and a compaction mould.

The erosion tests on compacted bentonite were conducted at 25°C and 60°C. The soil was compacted to the density of 1.4 Mg/m^3 at its natural water content of 12%. Deionised water was applied at the bottom of the cell with a constant flow rate of 0.46 mm^3/s. The volume of the outflow was approximately 40 mL/day. The experiments were at constant temperature which was monitored by the internal thermal sensor. The collected solution from the right bottom opening was used for dynamic lighting scattering (DLS) measurement. The DLS measurement was used to determine the concentration of the collected solution. There was no volume change during the whole process. In this case, the development of swelling pressure was monitored.

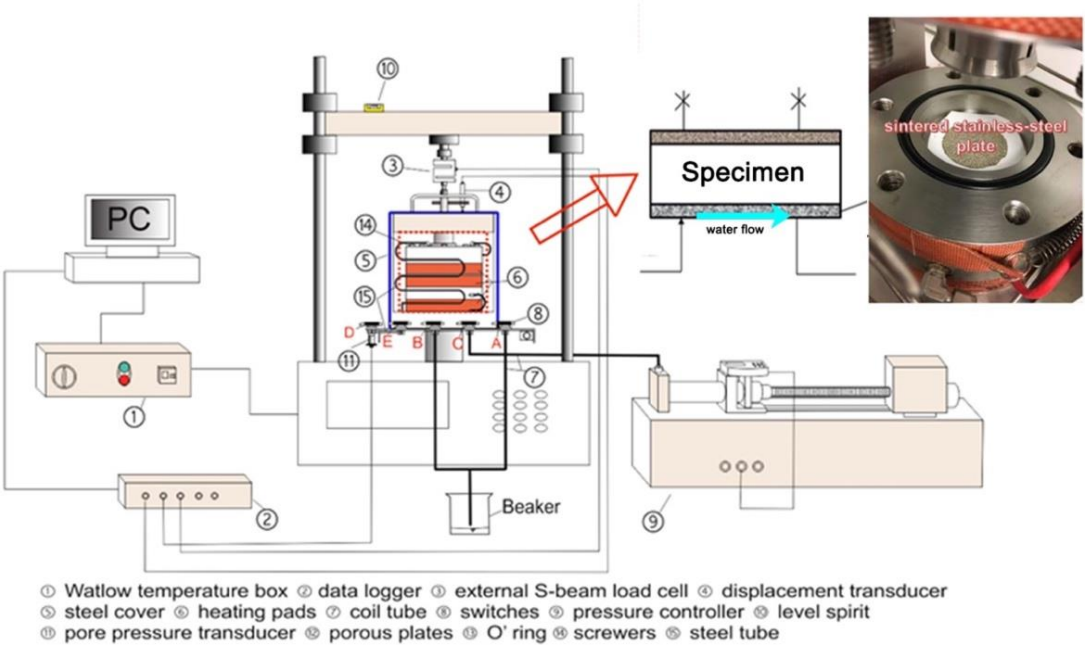


Figure 2. *Schematic diagram of the experimental setup.*

3 RESULTS

3.1. Erosion rate and mass loss

The water samples were collected at regular intervals and characterised by dynamic light scattering (DLS) by which the sizes and concertation of colloid particles were determined. A calibration curve was established by measuring the response of photomultiplier (cps, counts per second) at different and known concentrations of bentonite colloids. This enabled correlating between the laser counts in DLS measurement and bentonite concentrations from the calibrated curve. Each data point represents the average value of counts that were taken from the same water samples and measured by DLS three times. Figure 3 presents the variations of the colloid concentration with the total volume of water collected during the tests from the DLS results.

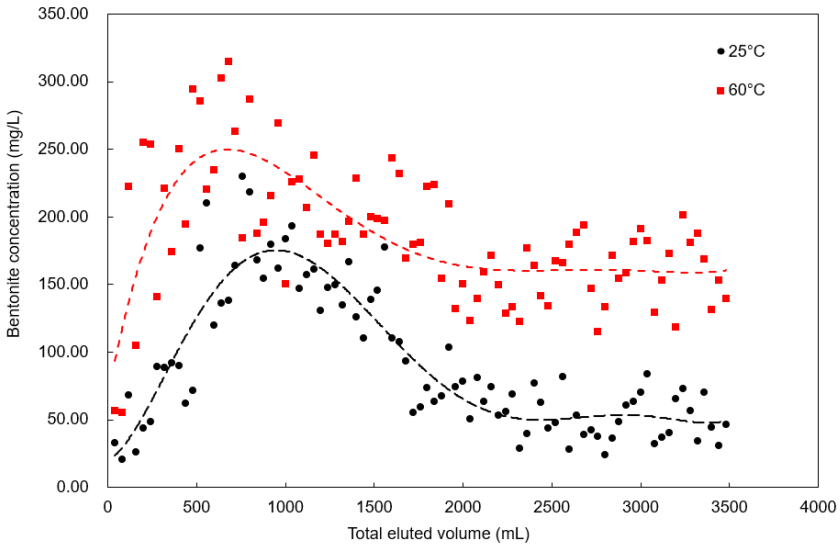


Figure 3. *Variations of concentrations of particles with the total eluted volume of water.*

The highest concentration among the daily collected samples was 320 mg/ L. After approximately 50 days, the system reached a steady state at high temperature. The time taken to steady state at low temperature was 60 days. The results show that the equilibrium concentration of colloidal solution was

higher at 60 ºC (i.e., approximately 50 mg/L at 25°C and approximately 170 mg/L at 60°C). Correspondingly, the eroded mass increased from 2 mg/day to 7 mg/day with increasing temperature. The total calculated loss masses were 0.315g and 0.643g for ambient and elevated temperatures, respectively. Accordingly, the mass loss was 2% and 4.1% of the initial mass.

In order to assess further the effects of temperature on erosion, the DLS tests were accompanied by turbidity measurement of the colloid solution (daily measurements). The results of turbidity measurements are presented in Figure 4. The trend observed in the results of turbidity measurements (NTU) is consistent with the behaviour observed in the DLS results. At steady state, the value of the turbidity fluctuated between 20 NTU and 30 NTU at higher temperature, and between 3 NTU and 5 NTU at 25°C.

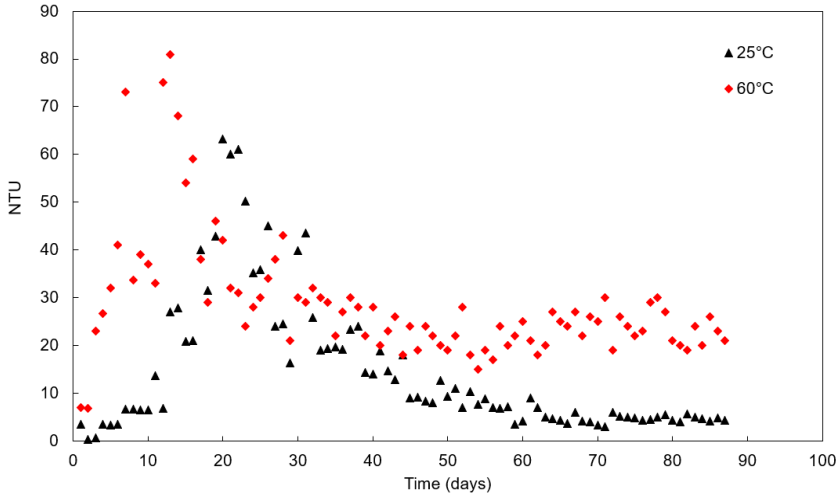


Figure 4. *Variations of NTU values of the turbidity measurements with time.*

3.2. Particle sizes of eroded mass

Figure 5 presents the results of DLS analysis of the particle sizes where the variations of the mean size of the colloid particles with total eluted volume of water are shown. The results show that the difference between the average particle size at 25ºC and 60ºC is small. The average particle size at the elevated temperature is smaller than ambient temperature but the differences are not significant. The analysis of particle size distribution for each solution did not provide notable trend, and it therefore acknowledged that further investigation is required to reveal the impact of temperature on size distribution of particles.

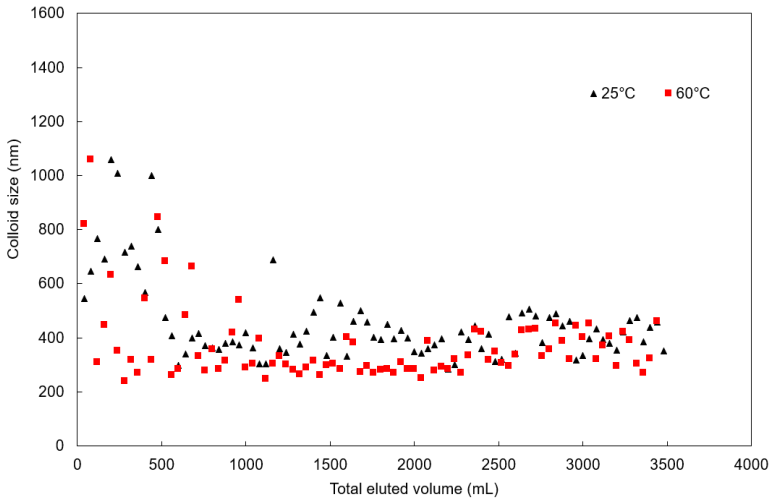


Figure 5. *Variations of the mean particle sizes with the total eluted volume of water.*

3.3. Swelling and extrusion

The specimen was partially confined, and the extrusion (swelling) of the sample was only allowed to happen within the steel porous plate. Due to the confinement of the sample from top, it was possible to measure the swelling pressure development at the top of the sample. The measurement of extrusion of the sample into the porous steel plate was not directly possible. However, the swelling pressure measured at the top of the sample, provides insights into potential impacts of temperature on extrusion. Figure 6 presents the evolution of swelling pressure of samples with time. It is noted that the water uptake has likely continued after reaching the peak which has resulted in continuous extrusion of clay to the porous stone. This is reflected in the drop of swelling pressure in Figure 6 which is more pronounced for the case of high temperature.

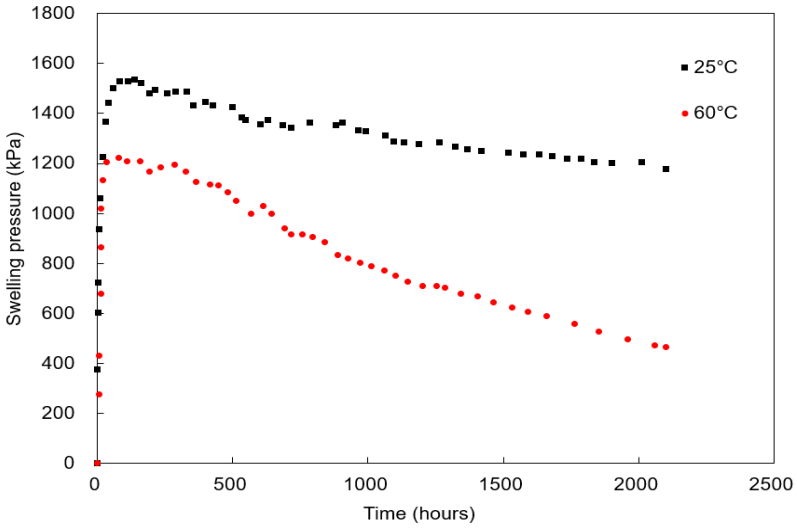


Figure 6. *Variations of the swelling pressure at the top of the samples with time.*

4 DISCUSSION

The discussions presented here intends to provide some preliminary insights about the complex processes and impacts that are generated by higher temperature. It is noted that the overall erosion for a given bentonite (including the three processes discussed in Figure 1) is dependent on various environmental and boundary conditions such as the flow rate and chemical composition of the eroding fluid. This study focused on providing the first observation at laboratory scale (to the best of our knowledge) on the effects of elevated temperature on erosion based on boundary and initial conditions that are not necessarily all represent the conditions of the repository. We acknowledge some of the conclusions cannot be generalised at this stage for other initial and boundary conditions (e.g. higher density, higher flow rate or chemical composition) and further investigations are required.

At the beginning of the experiments, the swelling pressure increased rapidly due to the water uptake. The rate of absorbing water is apparently larger than the rate of extrusion to the porous steel (bottom boundary). The swelling pressure reached a peak; then decreased continuously. The rate of swelling pressure reduction during this stage is larger at 60^{o}C than that of ambient temperature. The possible reasons for the higher erosion at 60^{o}C can be divided into two groups: (i) smaller initial extrusion of the clay to the porous boundary; and (ii) thermal effects on extrusion, collide generation, water properties and other intrinsic properties. In this study, the interface material at the bottom boundary at which the erosion has occurred (which is a porous medium) has important effects on how the overall erosion has evolved. At the bentonite-fractured rock interface the clay expand to the open fracture space and the erosion boundary moves with the clay. In this experimental study, the erosion occurs at a porous boundary, namely the porous steel.

Uptake of water would immediately result in the expansion and extrusion of the clay into the pores of the porous steel. For the case of high temperature, the swelling peak (1200 kPa) is smaller than that at the ambient temperature (1500 kPa). This suggests that a smaller quantity of clay was extruded into the pore space of the porous steel at the higher temperature. Therefore, larger voids were available for the

clay to form sol and to be eroded with the percolating water. On the other hand, there is another impact associated with the smaller extruded clay at higher temperature which is related to seepage velocity. The seepage water velocity (v_s) in the porous steel continuously decreases because the discharge velocity (v) is kept constant by the pump while the porosity (n) is reduced ($v_s = v/n$). The shear force that acts on clay particles due to water flow in pores increases with seepage velocity. Initially higher seepage velocity and sheer forces on the clay particle are expected for the case of erosion at ambient temperature as there is less solid extruded to the boundary during swelling (up to the peak value). However, the rate of the swelling drop is larger at higher temperature; therefore, larger amount of solid can enter to the porous space which increases the seepage velocity and shear forces on particles by reducing the porosity.

The stability of colloids (or yield stress) is reduced at higher temperature. Based on DLVO theory, the potential energy (G_{TOL}) between charged particles is the sum of the repulsion and the attraction components (Derjaguin, 1941):]

$$G_{TOL} = G_{VdW} + G_{DDL} \tag{1}$$

where, G_{DDL} is the repulsion electrostatic potential and G_{VdW} is the van der Waals attraction component; given by (García-García et al., 2006):

$$G_{VdW} = -\frac{A_H r_p}{12h} \tag{2}$$

and

$$G_{DDL} = \frac{64\pi r_p n_0 k_B T e^{-\kappa h}}{\kappa^2}\left(\frac{e^{\frac{ze\sigma^0}{2k_BT}} - 1}{e^{\frac{ze\sigma^0}{2k_BT}} + 1}\right)^2 \tag{3}$$

where, A_H is Hamaker constant, *h* is the separation distance between the particles, r_p is the radius of particle, n_0 is the number of background anions and cations, σ^0 is the surface charge density; k_B is the Boltzmann constant; z is valence of ions in the pore system and $\varepsilon_0\varepsilon_R$ is the dielectric constant.

Fig. 7 shows the results of DLVO calculations for potential energy at different temperatures. The maximum total potential energy (G_{TOL}) is the key factor to determine the status of the colloids. It can be seen that G_{DDL} decreases with temperature increase. The effects of temperatures on G_{VdW} can be neglected. G_{TOL} decreases accordingly with temperature and the reduction of the total potential decreases the stability of colloids. In general, this may lead to faster aggregation of colloids since the Brownian force is enhanced at higher temperatures and this contributes to the probability of particle collision and destabilizing the colloidal system.

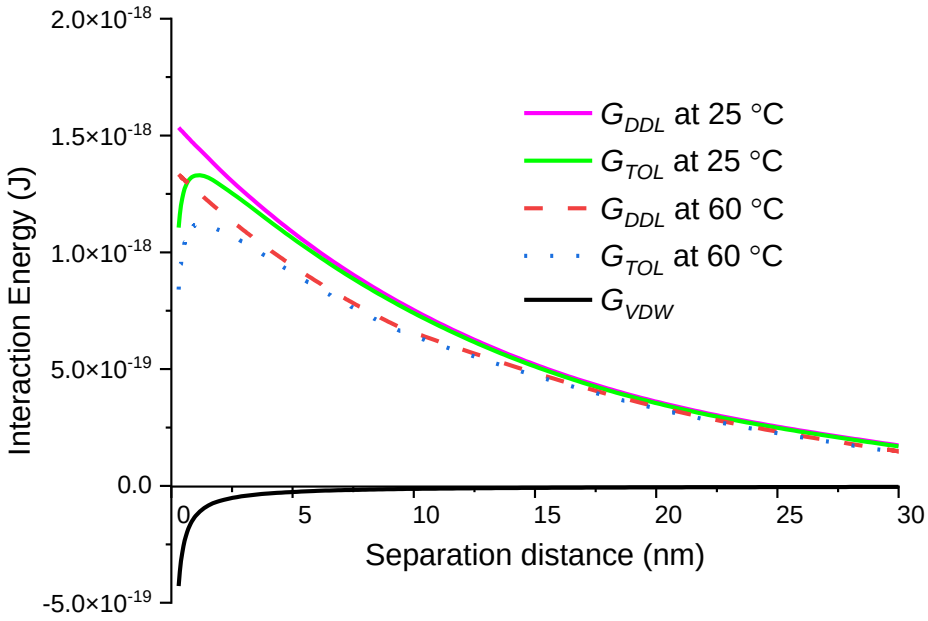


Figure 7. *Interaction energy as a function of separation distance at different temperatures*

5 CONCLUSIONS

This paper presented the results of laboratory experiments on the erosion of compacted bentonite at ambient and elevated temperatures. Compacted samples of Volclay bentonite (dry density 1.45 Mg/m^3) were subject to a constant flow of deionised water fat a porous boundary for three months. The results presented in this paper provides new insights into the erosion phenomena at elevated isothermal temperature in smectite rich clays. Our findings indicated that temperature effects on swelling and extrusion played a significant role in the overall erosion. The impact is partly due to the effects of boundary conditions of the experiment that is a porous medium due to the extrusion of clay. The lower swelling at higher temperature contributed to the increase of formations of clay gels and sol in the pore spaces of the porous steel which allowed the higher rate of erosion. Temperature was found to increase the overall colloid generation rate. The changes to the repulsion energy by higher temperature contribute to the stability of colloids. Further investigations are important to identify the impacts of high temperature on intrinsic erosion properties (e.g. rate of colloid generation and stability). The experimental system introduced in this paper provides a new capability to study the erosion in combination with other mechanical process.

6 ACKNOWLEDGEMENTS

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