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Preliminary Reporting of Thermophysical Property Measurements for the Ghareb Formation

Stephen Bauer and Charles Choens

Prepared by
Sandia National Laboratories
Albuquerque, New Mexico
87185 and Livermore,
California 94550

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ABSTRACT

Accurate knowledge of thermophysical properties of rock is vital to develop meaningful models of high level nuclear waste emplacement scenarios. The Israel Atomic Energy Commission is considering storing high level nuclear waste in the Ghareb formation, a porous kerogen bearing chalk. Sandia is supporting this effort with an evolving lab-based geomechanics testing program. We have completed measurements of thermal properties up to 275C and room temperature hydrostatic compaction measurements. We report thermal conductivity, thermal diffusivity, specific heat, and mass loss from our thermal measurements, and we report bulk moduli and porosity loss from our compaction measurements. These values are crucial for the numerical models to simulate heat transfer and formation compressibility around a heat generating repository.

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1. INTRODUCTION

This report describes preliminary measurements of thermal hydrological mechanical (THM) properties of the Ghareb formation in Israel, the target layer for geological disposal of high level nuclear waste. This effort is part of a collaboration which brings together unique experimental facilities from Sandia National Laboratories (SNL), and state-of-the-art computational capabilities from Geological Survey of Israel (GSI) and Lawrence Livermore National Laboratory (LLNL). The numerical study will utilize the state-of-the-art 3-D Hydro-PED numerical software recently developed in the Geological Survey of Israel, and the GEOS massively parallel multi-physics code under development at LLNL. Outcomes of this collaboration will include new and validated THM modeling capabilities and experimental data under geologic conditions relevant to radioactive waste disposal. The new capabilities developed will provide mutual benefit to both American and Israeli scientific communities.

Storing high-level nuclear wastes in geological formations provides a robust level of isolation of waste and has been embraced by all countries that have decided on a long-term waste-management strategy [Metlay, 2016]. The target layer for geological disposal in Israel is the Ghareb formation, a porous kerogen bearing chalk at a depth of about 500m. Designs for the geological disposal facility will combine a suitable system of engineered barriers with a favorable host rock that naturally has containment properties at sufficient depth that ensures adequate isolation from man and the environment. In a geological nuclear waste repository, the in-situ state of stress, temperature and hydraulic pressure in the host rock will be subjected to a few perturbations during installation and operation of the repository, including excavation of underground openings and thermal loading generated by radioactive decay from waste. Rock mass responses to these perturbations are coupled phenomena involving thermal (T), hydrological (H), mechanical (M), and chemical (C) processes [C-F Tsang et al., 2015]. Research has been conducted globally in several geological host formations to investigate the performance and safety of potential underground nuclear waste repositories. These include crystalline rock, indurated and plastic clays, and domal and bedded salt [Ewing et al., 2016]. Several European countries consider clay formations well-suited to hosting radioactive waste repositories because of their very low hydraulic conductivity and self-sealing capacities [Bastiaens et al., 2007; C F Tsang et al., 2012]]. Rock salt is another potential medium characterized by low primary permeability, low porosity, high ductility, healing capacity, and relatively high thermal conductivity [Hunsche and Hampel, 1999]. Sweden and Finland are considering potential repositories in granitic rocks due to their high mechanical strength, low porosity, and paucity of fractures [Hedin and Olsson, 2016]. Ultimately, host formation selection is driven by availability and socio-economic factors in individual countries.

The excavation of deep-buried underground openings and heat generated by decay of radioactive wastes after emplacement may induce damage and progressive failure of surrounding rocks. The ability of damaged rocks to provide preferential pathways for groundwater flow and radionuclides migration is an important concern for the long-term safety of a nuclear waste repository [Armand et al., 2014]. The coupled thermo-mechanical (TM) behavior of surrounding host rocks has gained increasing research interest for predicting safe disposal of radioactive waste. Understanding the response of the rock mass to excavation and thermal loading, therefore, is of paramount importance for safety assessment and optimal design of a nuclear waste repository. The focus of this work is on the mechanical damage that could occur within host rocks as a result of excavation and heating. To comply with safety requirements, the performance of a nuclear waste repository must be evaluated in the long-term (typically, thousands or even millions of years for heat-generating nuclear waste). Due to the complexity of the processes that need to be investigated, their interactions and the time scales considered, numerical modeling using proper tools and state-of-the-art knowledge is required [Blanco Martín et al., 2015].

The Israeli Atomic Energy Commission is shifting repository consideration from the initial deep borehole repository in the Zenifim formation (due to high cost of a 3-5 km deep borehole) to a shallow (500 m) borehole disposal in an organic rich high porosity carbonate rock, the Ghareb formation. The new formation presents a new set of technical challenges which we are assisting to identify and address. The new direction/motivation of this collaborative project is to evaluate the scientific and safety case for intermediate borehole disposal of Israel's nuclear waste. The new formation requires an evaluation of the long-term security and safeguards in a sealed borehole at shallower depths and an evaluation of the long term isolation capacity of the Ghareb formation for radionuclides. The IAEC will focus on evaluating the feasibility and subsequent implementation of geologic disposal of nuclear waste in borehole(s), including drilling and borehole design, waste handling and downhole emplacement, waste package design, borehole seal design, and development of a safety case for operations and long-term waste isolation and storage. We have begun THM characterization of this new target horizon, the Ghareb formation, and we report on initial measurements on thermal properties and hydrostatic compaction behavior.

The Ghareb formation is up to 120 m thick in the western parts of the Yasmin Plain, and is composed of chalk and marl, oil shales, with kerogen (Bisnovat et al., 2015, Shitrit et al., 2016) and relatively high-porosity (20-45%). The samples used in this study were prepared from blocks collected by the authors from a quarry.

2. TESTING SETUPS AND METHODS

2.1. Specific Gravity

Specific gravity is the density of a substance divided by the density of water. Since water has a density of 1 gram/cm^3 (at standard temperature and pressure conditions), and since the units cancel, specific gravity is usually very close to the same value as density (but without any units). Specific gravity measurements are made by grinding the rock to a powder, drying at a specified temperature, weighing it, and then measuring the sample volume by liquid displacement.

2.2. Thermal Properties

Thermal property measurements, namely, thermal conductivity, diffusivity, and specific heat, were performed on 5 randomly chosen samples from rock collected at a surface quarry. Thermal measurements were made at ambient pressure and moisture conditions using a transient method based on the theory of the transient plane source technique with a Hot Disk Thermal Constants Analyzer (TCA). The transient plane source method uses a planar, Kapton-coated nickel coil that functions as both a heat source and resistance thermometer capable of detecting temperature changes as slight as 10^{-4} K. The samples used are well above the minimum TCA sample size of 3 mm in height by 7 mm in diameter; there is no maximum sample size. The rock samples were cut with a diamond saw and opposing surfaces were ground smooth with a 400 grit lap wheel. The TCA coil is sandwiched between the two halves (Figure 1) using less than 5 psi mounting pressure. Passing a current through the coil increases its temperature by as much as a few degrees Celsius and causes a pulse of heat to enter the sample. The coil temperature then decreases over time as a function of the sample's ability to conduct heat away from the coil. By monitoring the coil temperature over time, the TCA simultaneously calculates sample thermal conductivity, thermal diffusivity and specific heat .

Two different temperature profiles were used to measure thermal properties. In the first set of measurements, samples were placed on the test system and the temperature was cycled up to the maximum temperature, pausing every 25C to make measurements. Measurements were made for both increasing and decreasing temperature ramps. For the second set of measurements, sample mass was recorded at each temperature to determine the amount of water loss at each temperature interval. At each temperature the sample equilibrated for a few hours before a measurement was made. Measurements were made at room temperature and at 50C intervals up to 275C.

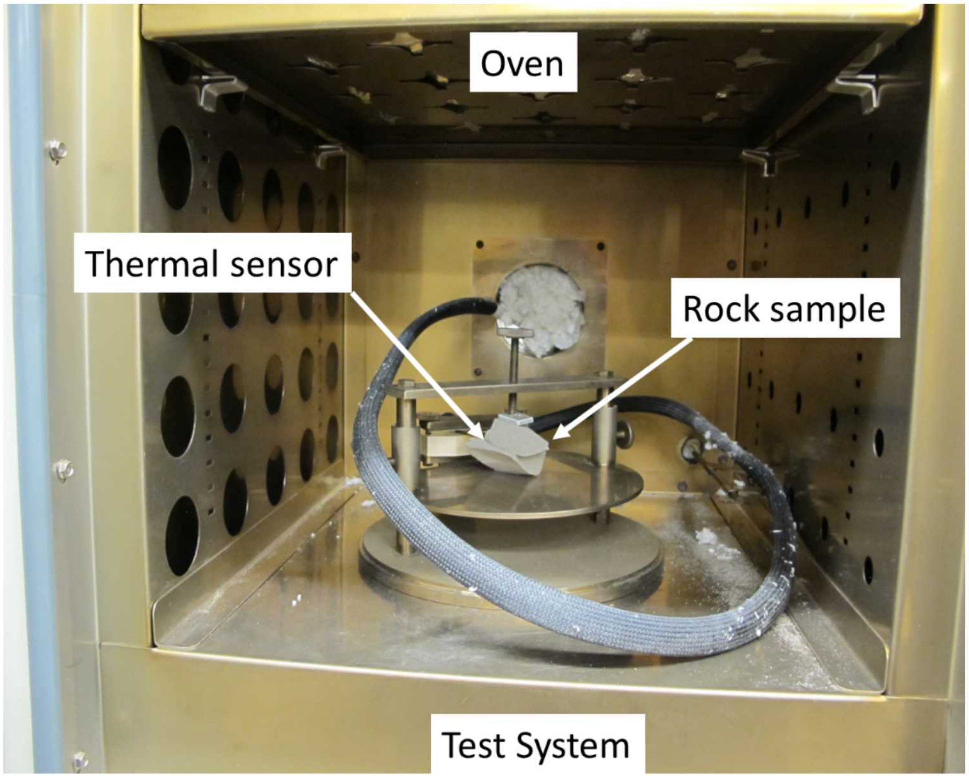

Figure 1 Thermal Measurements Test System.

2.3. Bulk Modulus

Bulk Modulus was determined through hydrostatic compaction of a sample to 400 MPa. The sample was 3.8 cm diameter by 5.3 cm in length (Figure 2). The sample was jacketed in 3 layers of paint on urethane jacketing material and instrumented with two axial and two lateral LVDTs (transducers used to record displacements during application of pressure). Experiments were conducted in a pressure vessel with capacity of 400 MPa hydrostatic pressure and mounted in a servohydraulic load frame with a maximum capacity of 4.5 x 10^6 N. For the experiment, hydrostatic pressure was applied in steps with smaller unload/reload loops up to a specified maximum pressure. The sample was vented to the atmosphere to prevent any potential pressurization of internal fluids. Pressures and displacements were recorded by a digital data acquisition system that also controlled the frame. Unload/reload loops are completed to capture the elastic property (bulk modulus) after the material has experienced a certain amount of volume strain. From axial and lateral displacements, axial and lateral strains are calculated, and ultimately volume strain is determined. Volume strain was calculated from:

$$\varepsilon_V = \varepsilon_A + 2 * \varepsilon_L \quad (1)$$

where ε_A is axial strain and ε_L is lateral (radial) strain.

Figure 2 Hydrostatic test sample, note shortening of compacted sample.

3. TEST RESULTS

3.1. Specific Gravity

Specific gravity measurements are given in Table 1. For two of the samples, drying temperature was 60°C, and for the third sample, it was 110°C; perhaps more water was removed from this last sample, yielding a greater specific gravity, and or it reflects slight changes in mineralogy with different grain densities. Also, the data set is very limited, so comments beyond general observations may be premature. The specific gravity of the Ghareb is low compared to other rock, for example granitic rock is on the order of 2.75, sandstone 2.7, shale 2.5, and limestone 2.6.

Table 1.Specific gravity determinations

Measurement	Sample weight (g)	Dry weight (g)	Gravimetric Water Content (%)	Temperature in the oven (Celsius)	Specific Gravity
1	57.73	51.33	12.5	60	2.01
2	70.79	60.73	16.6	60	2.17
3	7.67	6.48	18.4	110	2.21

3.2. Thermal Properties

Thermal property determinations, thermal conductivity, thermal diffusivity, and specific heat are presented in Figures 3, 4, and 5 , respectively. For each of the five samples used in the first temperature profile, temperature was ramped up and then down. The variations from measurement to measurement are small for any/all samples, the slight variations observed reflect the fidelity of the measurements.

Thermal conductivity (Figure 3) is in the range of 0.35 to 1.1 w/mK over the measured temperature range; most of the variation in values are due to sample to sample variations. Individual samples vary 0.1 to 0.2 w/mK for the entire temperature range. In general, the thermal conductivity decreases with increasing temperature, except for one sample, which increases slightly. There is little to no hysteretic behavior observed upon cooling.

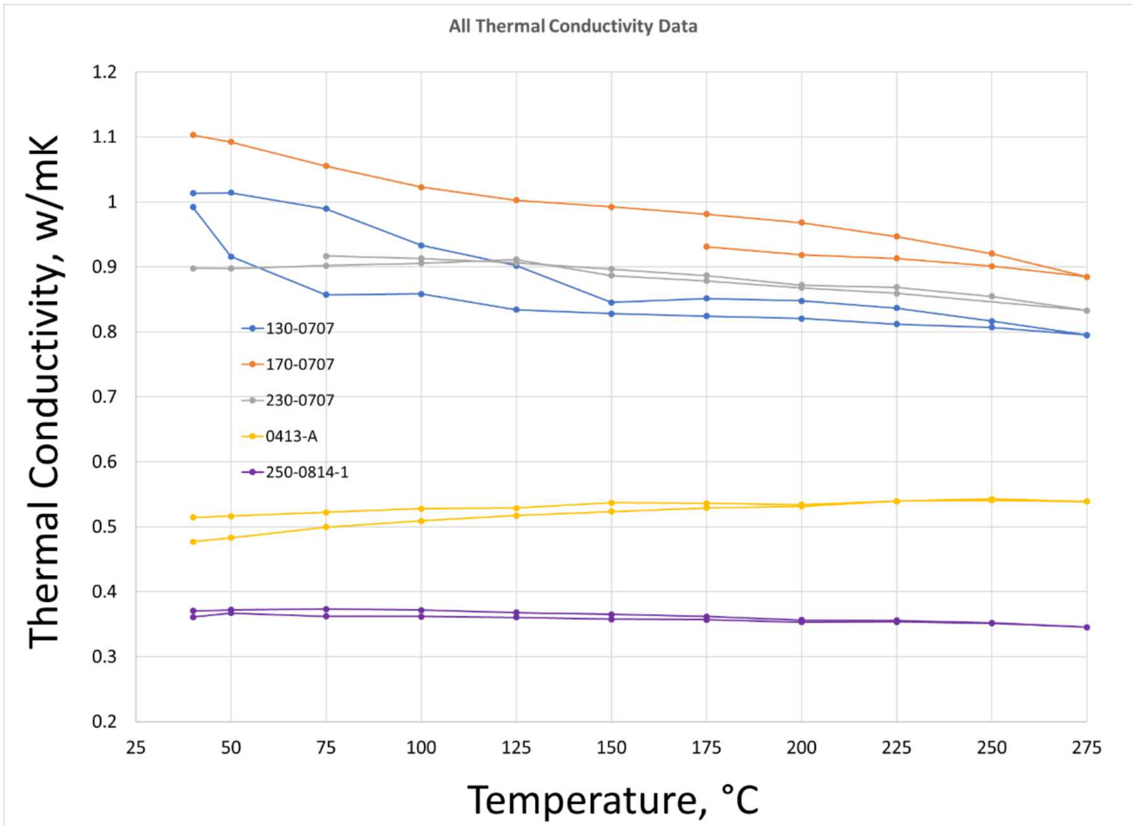

Figure 3 Thermal conductivity versus temperature

Thermal diffusivity (Figure 4) is in the range of 0.27 to 0.7 mm^2/s over the temperature range with most of the variation caused by sample to sample variations. Individual samples vary 0.1 to 0.2 mm^2/s for the entire temperature range. The thermal diffusivity decreases with increasing temperature for all samples. There is little to no hysteretic behavior observed upon cooling.

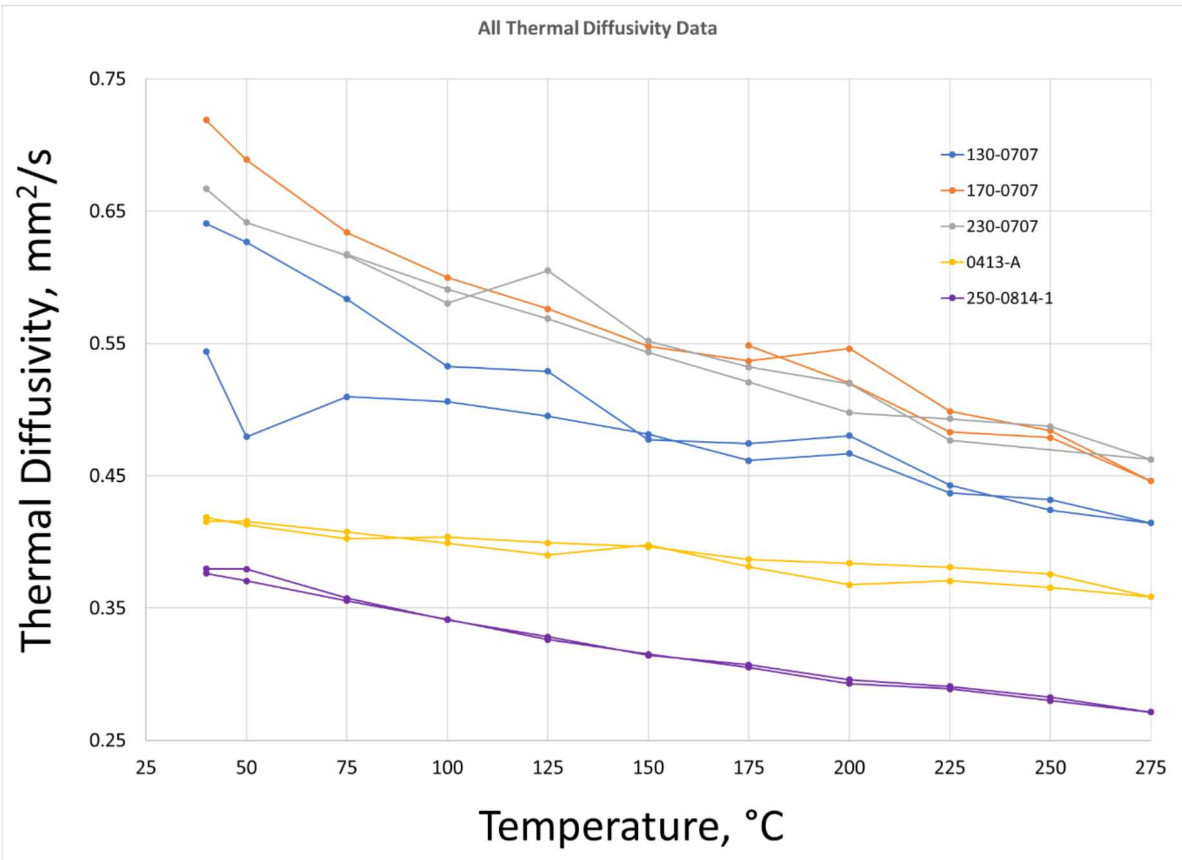

Figure 4 Thermal diffusivity versus temperature

Specific heat (Figure 5) is in the range of 0.95 to 2.0 MJ/m^3K over the temperature range with most of the variation caused by sample to sample variation. Individual samples vary 0.2 to 0.4 MJ/m^3K for the entire temperature range. The Specific heat increases with increasing temperature for all samples. There is little to no hysteretic behavior observed upon cooling.

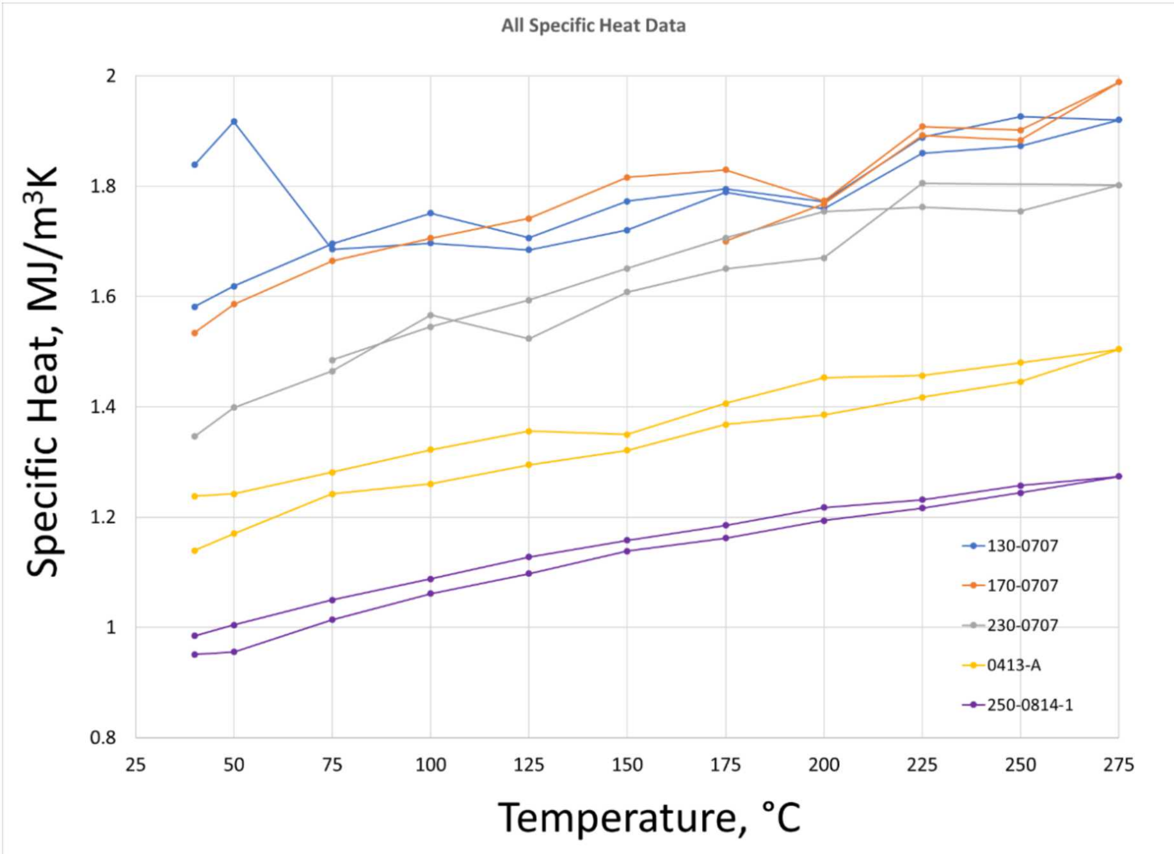

Figure 5 Specific heat versus temperature

A final sample was run with the second temperature profile, where the sample was cooled to room temperature and removed from the test system for mass determination in between temperature steps to determine volatile loss (Figure 6). Potential volatiles in the Ghareb formation are water and organic matter, which could be detected via its characteristic odor upon heating to temperatures above about 250 °C. Volatiles were continuously lost from ambient conditions through about 125 °C, after which the mass remained roughly constant to about 200 °C, and from 200 °C to 275 °C, more volatiles were released. We interpret the low temperature loss to be water, and the high temperature weight loss to be organics.

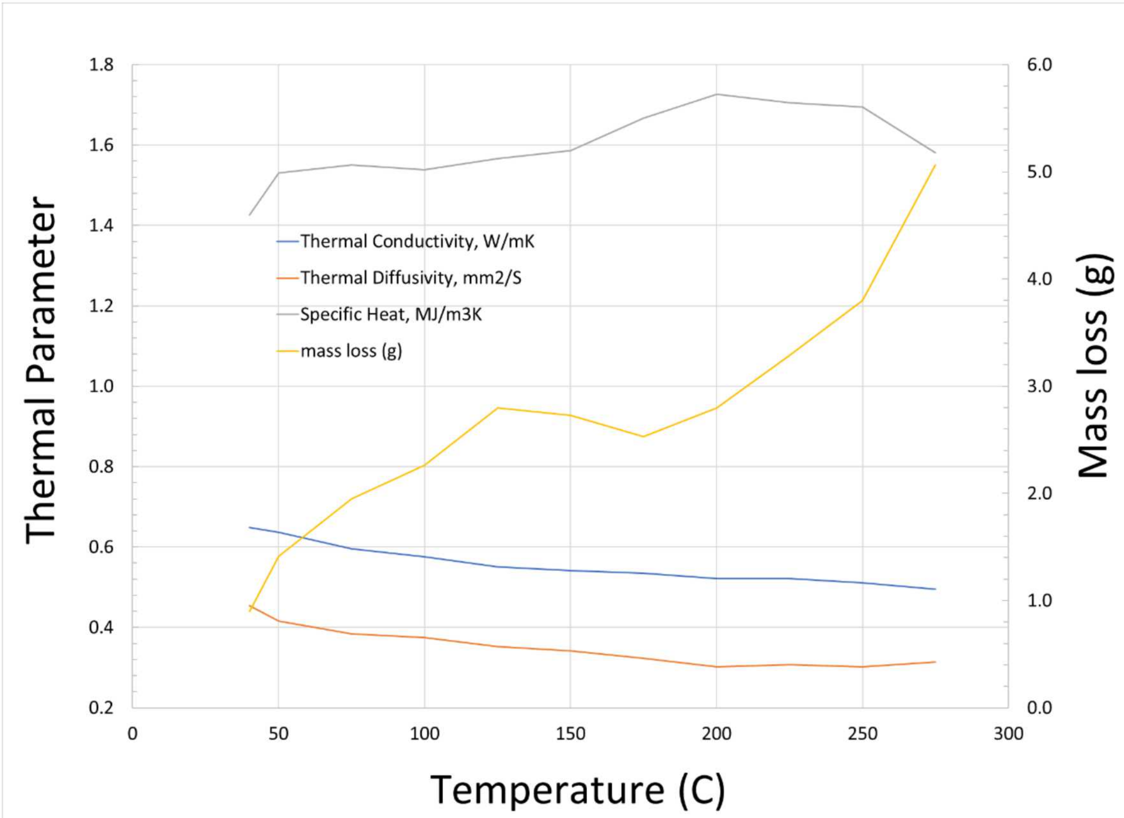

Figure 6 All thermal properties and weight loss versus temperature

3.3. Bulk Modulus

Bulk modulus is determined from the relationship between pressure and volume strain in a hydrostatic compaction test (Figure 8). In this reporting, the linear slope of the pressure-volume strain relationship is used to represent the bulk modulus. From this plot it is apparent that the material stiffens with increasing pressure (note increase in slope), and most strain is permanent, with only 5% recovered after compaction to 400 MPa.

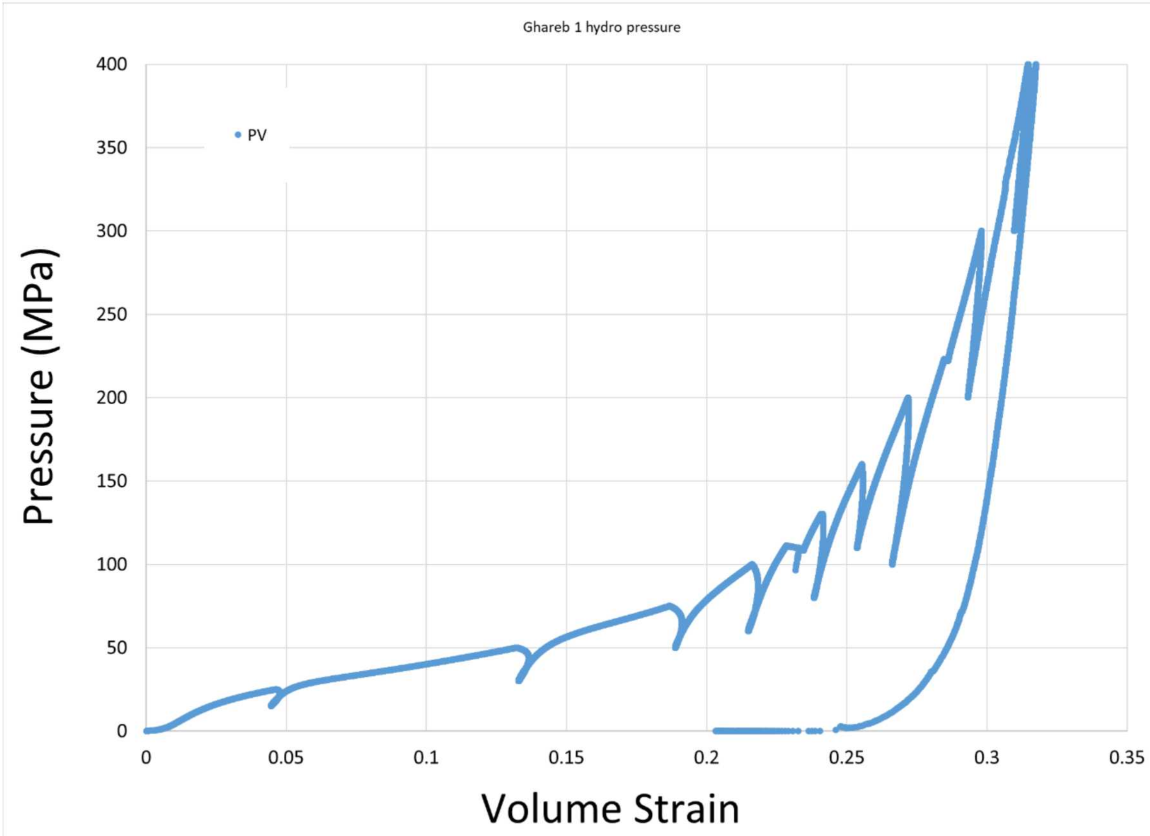

Figure 7 Volume strain versus pressure

The bulk modulus (K) is plotted versus pressure in Figure 8. The plot shows 2 pressures for each modulus. The blue dots present the average pressure of the measurement range, and the orange dots present the maximum pressure of the measurement. It is also important to note that at 350 and 400 MPa, K remains near constant, suggesting that most of the porosity has been consolidated from the sample.

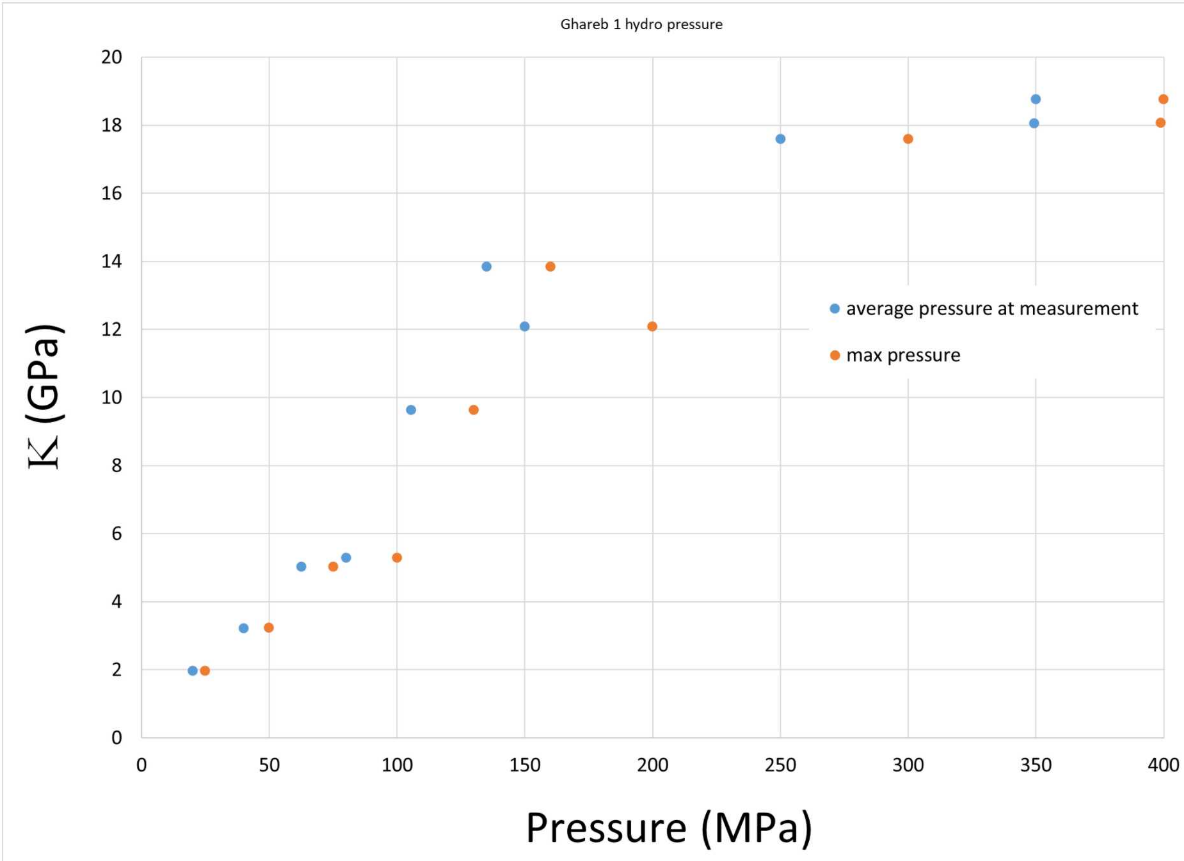

Figure 8 Bulk Modulus versus pressure

Density increases while porosity greatly decreases with increasing pressure (Figure 9). As we are measuring volume strain, and mass of the sample is constant, density is calculated by the dividing the starting density of the sample by the evolving sample volume during the test. Next, the porosity is calculated using well known relationships between the current density and bulk density. At maximum pressure, the sample approaches near zero porosity, and similarly, the sample density approaches the grain density determined separately.

Lastly, we present a relationship between density and bulk modulus demonstrating that the bulk modulus increases with increasing density (Figure 10). The relationship can be described with a 2 linear slope relationship. The initial density-bulk moduli curve is steep during initial compaction, but as porosity is reduced, the slope of this relationship is much lower. The change in slope occurs around bulk moduli values of 6-8 GPa, corresponding to ~100 MPa hydrostatic pressure. This relationship is important for modeling of the material as is compacts.

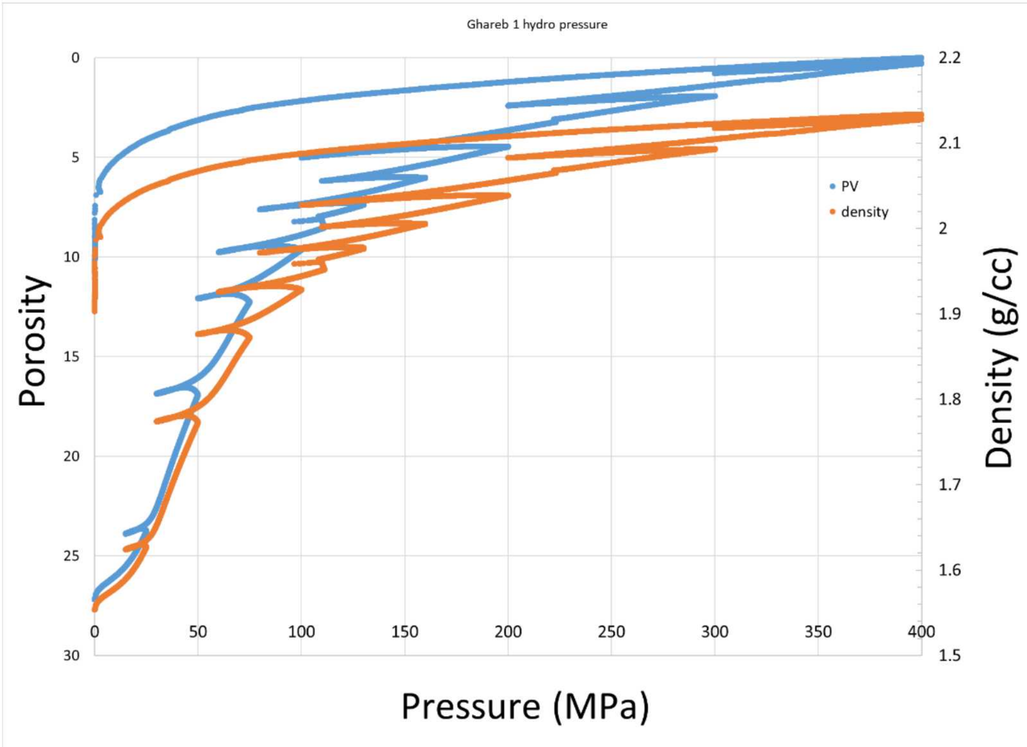

Figure 9 Porosity and density versus pressure

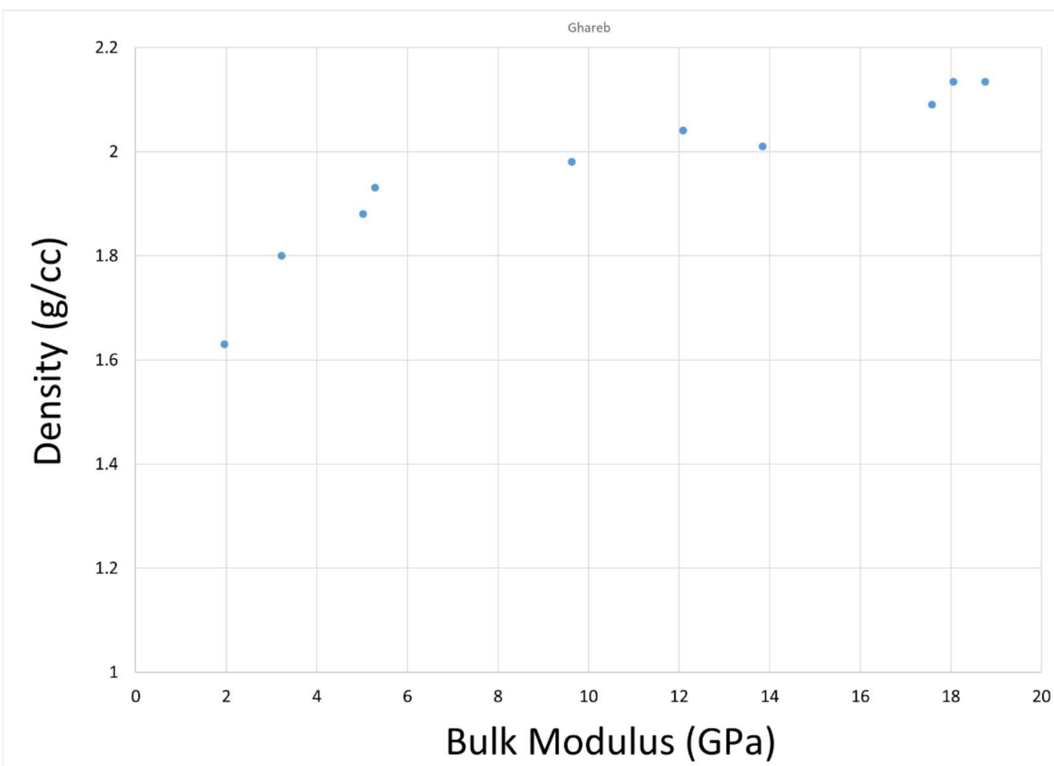

Figure 10 Density versus bulk modulus

4. SUMMARY

A preliminary reporting of thermophysical property measurements for the Ghareb formation has been completed. The grain density is on the order of 2.13, much lower than many other sedimentary rocks, reflecting mineralogical, microtextural, and potential organic content of this rock compared to others. A set of thermal properties has been determined, and for each parameter up to 275°C, there is great sample to sample variability and little hysteretic behavior. Finally, a single hydrostatic pressurization experiment up to 400 MPa reduced porosity to near zero and increased sample density to almost grain density while demonstrating that the bulk modulus is extremely pressure sensitive. We will use these tests and published literature to guide us in our future geomechanical test planning to best parameterize numerical simulations to establish safety cases for an intermediate depth borehole repository in the Ghareb formation.

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