MASTER

UCRL-83789, Rev. 1
PREPRINT

CONF-791134--12(Rev 1)

THERMAL CONDUCTIVITY, DIFFUSIVITY, AND EXPANSION OF AVERY ISLAND SALT AT PRESSURE AND TEMPERATURE

W. B. Durham
A. E. Abey
D. A. Trimmer

This paper is being published in the
Proceedings of the 16th ITCC/7th ITES Conference, Chicago, Illinois
November 7-9, 1979

March 4, 1981

Lawrence Livermore Laboratory

THERMAL CONDUCTIVITY, DIFFUSIVITY AND EXPANSION OF AVERY ISLAND SALT AT PRESSURE AND TEMPERATURE

W. B. Durham, A. E. Abey, and D. A. Trimmer

Lawrence Livermore National Laboratory
P.O. Box 808
Livermore, CA 94550

ABSTRACT

Preliminary data on the thermal properties of a coarse-grained rock salt from Avery Island, Louisiana, indicate that hydrostatic pressure to 50 MPa has little effect on the thermal conductivity, diffusivity and linear expansion at temperatures from 300 to 573 K. The measurements were made in a new apparatus under conditions of true hydrostatic loading. At room temperature and effective confining pressure increasing from 10 to 50 MPa, thermal conductivity and diffusivity are constant at roughly 7 W/mK and 3.6×10^{-6} m^2/s, respectively. At 50 MPa and temperature increasing from 300 to 573 K, both conductivity and diffusivity drop by a factor of 2. Thermal linear expansion at 0 MPa matches that at 50 MPa, increasing from roughly 4.2×10^{-5}/K at 300 K to 5.5×10^{-5}/K at 573 K. The lack of a pressure effect on all three properties is confirmed by previous work. Simple models of microcracking suggest that among common geological materials the lack of pressure dependence is unique to rock salt.

INTRODUCTION

Prediction of the short- and long-term thermomechanical behavior of an underground repository designed for the storage of radioactive wastes strongly depends on the thermal response of the rock at the physical and chemical conditions <u>in situ</u>. A laboratory experiment has been designed to make thermal properties measurements of large-sized samples under conditions similar to the pressure, temperature, and hydrologic conditions which might be expected in a nuclear waste repository. We report here preliminary data taken on the first rock type tested, a coarse-grained domal rock salt from Avery Island, Louisiana.

The interest in isolating radioactive waste is obvious. Burying waste in the earth would be a straightforward means of isolation were it not for the fact that most rocks, being brittle materials, tend to crack under an applied load. If rock around a waste repository is cracked, the isolation condition may be violated: the permeability of the rock could increase and allow interchange of radioactivity with the biosphere. All rocks in the upper few kilometers of the earth's crust, in fact, contain some fractures, and the heat load imposed by radioactive waste could be expected to cause further cracking, especially on the microscopic scale, owing to local mismatches in thermal expansivity. Microcracking can also be induced (even in thermomechanically isotropic materials) in the presence of a sufficiently strong thermal gradient. Hydrostatic pressure is expected to suppress fracturing and work to close existing fractures. Studies of other crack-related phenomena in rocks such as porosity and elastic behavior (see Walsh and Brace, 1966, for a review of early work) indicate that the pressure effect on cracks tends to be more pronounced at low pressures. As more and more cracks close with increasing pressure, the effect becomes more subtle. Crack effects become unimportant above pressure of 10 to 300 MPa, depending on rock type.

Microcracking can affect thermal properties as well as the mechanical properties and experimentation has so demonstrated (Hasselman, this volume). Pressure, by its effects upon microfractures, is expected to affect thermal properties (Walsh and Decker, 1966), but experimental quantification of this effect, under conditions of true hydrostatic loading in the upper crust pressure range, is lacking.

Rock salt has been considered as a potential waste repository material primarily because of its high thermal conductivity (as measured at ambient pressure) and its relatively (to other rocks) high ductility which allows it to retain low permeability to fluid flow under loading conditions which would produce cracking in most ordinary rocks. In addition, rock salt stands out from other geological materials in extended geological formations in that it is composed of a single, thermomechanically isotropic phase. Differential thermal expansion, hence microfracturing due to temperature change, would not be expected to occur except in strong thermal gradient. Again, however, experimental confirmation is lacking.

EXPERIMENTAL TECHNIQUES

The experiment is designed to approximate heat flow from an infinite line source. Thermal conductivity (k) is determined by establishing steady state heat flow from the line source, then measuring temperature (T) as a function of distance (r) from the infinite line source, measuring the power per unit length of the line source (q/ℓ) and applying the axisymmetric heat flow law:

$$q/\ell = \frac{2\pi k}{\ell n(r_2/r_1)}(T_1 - T_2) \qquad (1)$$

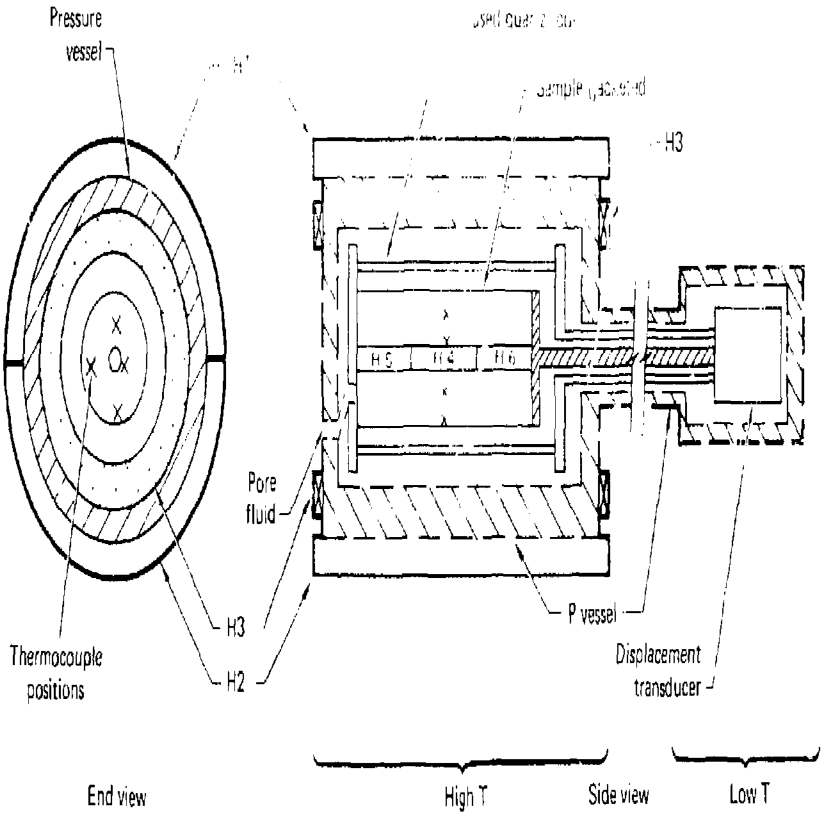
where $T_1 = T(r_1)$ and $T_2 = T(r_2)$. Thermal diffusivity (κ) is determined by pulsing the line source, measuring $T(r,t)$, where t is time, and applying the relationship

$$\frac{\partial T}{\partial t} = \kappa\left(\frac{1}{r}\frac{\partial T}{\partial r} + \frac{\partial^2 T}{\partial r^2}\right) . \qquad (2)$$

The pulse in this case is a step increase in power of the line source. Equation (2) is solved by an iterative approach using a digital computer (Abey, 1979).

A schematic of the apparatus is shown in Fig. 1 and a detail of the sample assembly in Fig. 2. Figure 3 is a photograph of the components of the sample assembly. An externally heated pressure vessel is used to set the pressure and temperature conditions for each test. The pressure medium is argon and the vessel is designed to sustain pressures up to 200 MPa (corresponding to burial depths of 6-7 km depending on rock type) at temperatures to 773 K. Although it is not employed for tests on rock salt, the means exist to introduce pressurized fluid into the sample to simulate <u>in situ</u> hydrologic conditions. Located at a remote "cold" point within the pressurized volume is a displacement transducer (LVDT) used to measure linear thermal expansion. Concentric stainless steel tubes connect the LVDT body and core rigidly to either end of the sample.

The 0.13-m diameter × 0.23 m long sample is sealed in a 0.25-mm-thick copper jacket to exclude the pressure medium from the rock. Were it not for the jacket, the pressure in the cracks and pores of the rock would equal the pressure outside, so that the effective pressure acting to close cracks and generally hold the rock together would be zero. Thermocouples and plumbing for the pore pressure fluid enter the sample through high-pressure seals. All thermocouples are type J (iron-constantan) and are clad in 1.6-mm-diameter stainless steel sheathing. The infinite line source condition is established by the arrangement of heaters



Sample size: 13 cm dia X 23 cm long

Operating limits: 773 K, 200 MPa

H1, H2, H3 – external heaters

H4, H5, H6 – internal heaters

Fig. 1. Schematic of the high-pressure thermal properties apparatus.

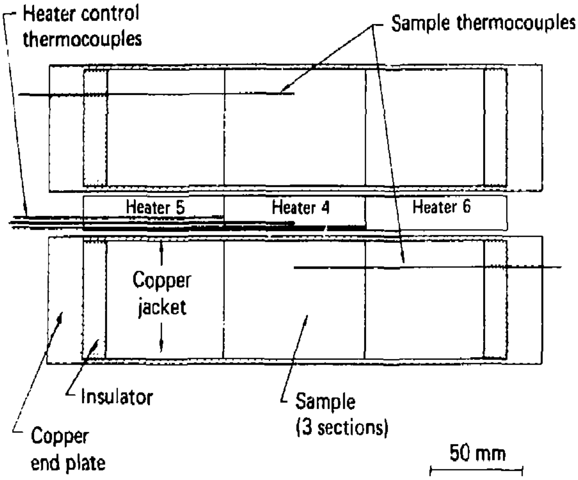


Fig. 2. Cross section of sample assembly for high-pressure thermal properties experiment. There are eight thermocouples in the mid-plane of the sample, four entering from each end. The tips lie at eight different radii in a spiral arrangement around the sample axis.

and control thermocouples shown in Fig. 2. The heat source is three separate resistance windings placed inside a single 3-mm-thick copper support tube (not shown in the figure) which itself is positioned in a 19.1-mm-diameter hole cored along the axis of the sample. The copper sample jacket, of course, lines this core and stands between the core heaters and the sample. The sample is sectioned to match the sectioning of the core heaters, and three control thermocouples are positioned at the center and at the ends of the central heater/sample section. The control thermocouples are silver-brazed to the outside of the copper heater tube. Power to the core heaters is supplied so as to hold the three control thermocouples at the same temperature. Since a zero gradient then exists along the axis of the middle section, it is presumed that all power delivered to the middle heater section flows into the middle section of the sample. A numerical simulation of the heat flow pattern in the sample, in the situation where the entire length of the core is held at constant T, indicates that of the heat which flows radially into the center section of the sample, less than 5% is lost through the ends of that section in the worst case ($T_{max} - T_{min} \approx 50$ K).

The most likely source of systematic error in the measurement of thermal conductivity and diffusivity is deviation from the presumed heat flow pattern. The experimental configuration is

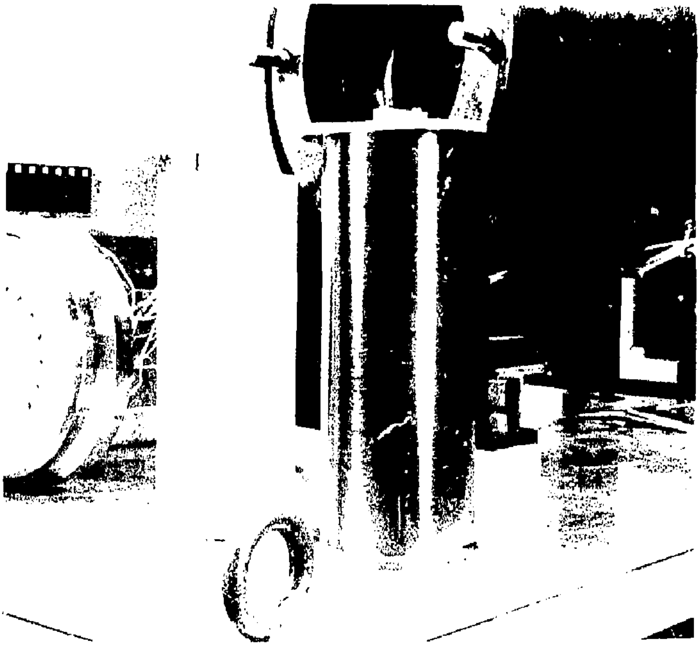


Fig. 3. Photograph of the sample assembly. The salt sample shown here stands 0.23 m high × 0.13 m in diameter and is cut into three equal sections. The large grain size (5-10 mm) of the salt is evident. Ceramic insulators are placed at either end of the sample and the arrangement is sealed inside a copper canister as shown at the right. In the background can be seen one end of the pressure vessel and a portion of the pressure sealing plug with its various power and signal feedthroughs.

complex and there are numerous ways such deviation could arise. Evaluation of systematic error is ongoing but is not complete. Random error from inaccuracies in the temperature and power measurement and from inhomogeneity of the sample (causing local temperature deflections) is expected to be small compared to errors in the system. We plan to approach the error analysis both empirically using standard materials and analytically using numerous heat flow monitoring thermocouples.

RESULTS AND DISCUSSION

The sample material was supplied by RE/SPEC, Inc., Rapid City, South Dakota. The rock has a porosity of approximately 1% and is unusually clean in appearance. Grain size averages 7.5 mm and ranges from 2.5 to 1.5 mm. The only impurities are a few anhydrite crystals and clay-like particles at grain boundaries. A chemical analysis of Avery Island salt given by Kaufmann (1960) shows roughly 99.1% NaCl, 0.7% water insolubles, and 0.2% $CaSO_4$. Water content is 0.02%.

A single sample has been tested to date. At room temperature, thermal conductivity and diffusivity appear to be independent of pressure to 50 MPa (Fig. 4). The error bars show one standard deviation of repeated measurements (20 to 40), so are an indication only of random error. No explanation is offered for the anomalously high diffusivity at 10 MPa. As a function of temperature to 573 K at a constant pressure of 50 MPa (Fig. 5), conductivity and diffusivity both show a monotonic decrease with increasing temperature. Linear expansion was measured from room temperature to 573 K at two pressures: 0 and 50 MPa (Fig. 6). The coefficient of linear expansion may show a slight decrease with confining pressure. Throughout the experiment temperature gradients in the sample were kept low to avoid gradient-induced cracking. The maximum steady-state gradient for the conductivity measurement was 3 K/50 mm.

The data are compared to other published values in Figs. 6 and 7. The vast majority of these values are based on an extensive compilation of values by Yang (1981). In general, there is good agreement between our values and previously measured values, despite the preliminary nature of our measurements. The

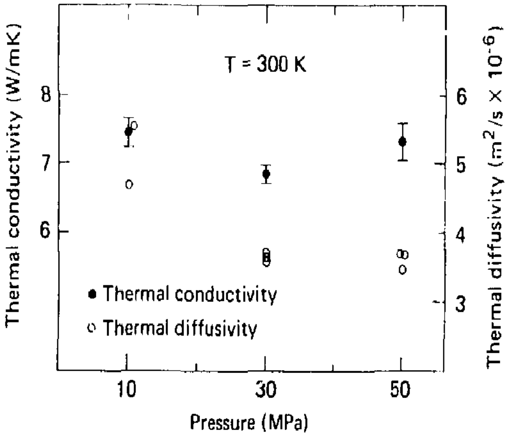


Fig. 4. Thermal conductivity and diffusivity of Avery Island rock salt as a function of confining pressure measured near room temperature.

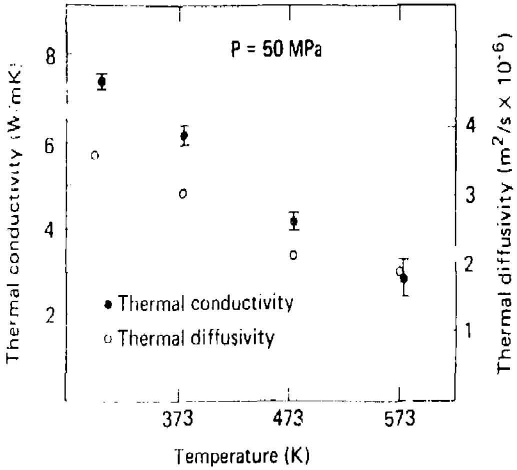


Fig. 5. Thermal conductivity and diffusivity of Avery Island rock salt as a function of temperature measured at 50-MPa confining pressure

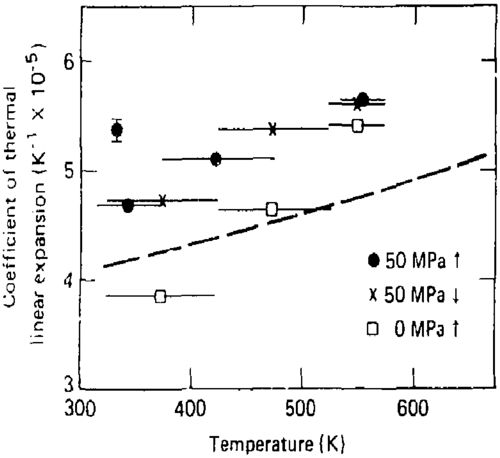


Fig. 6. Thermal expansivity of Avery Island rock salt as a function of temperature and pressure on heating (indicated by up arrow) and cooling (down arrow) cycles at 0- and 50-MPa confining pressure. Each point is the average coefficient of linear expansion over the temperature range indicated by the horizontal bars. The vertical bar shows the estimated error of the average. The dashed line is from Yang (1981).

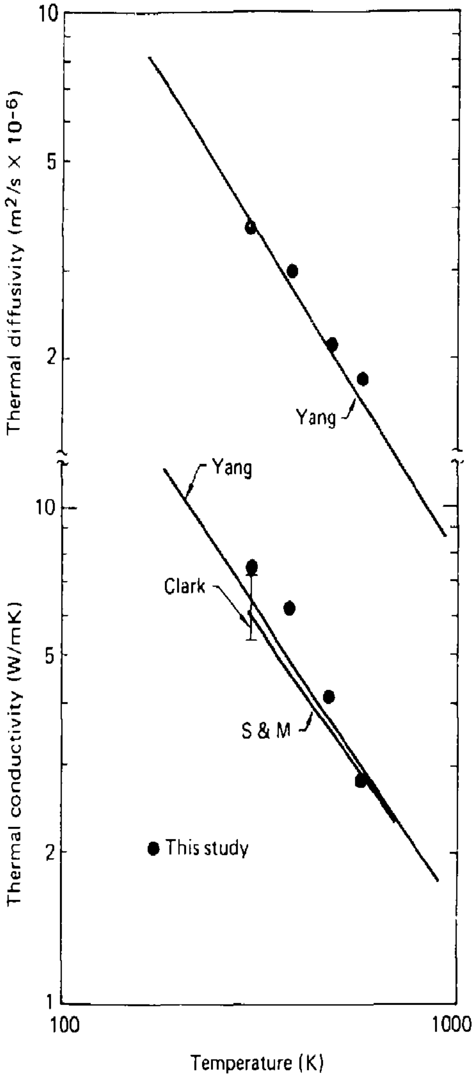


Fig. 7. Conductivity and diffusivity compared with previously measured values. The solid lines from Yang (1981) are the CINDAS "recommended" values and represent measurements on single crystals and polycrystals at very high confining pressures. The line from Sweet and McCreight (this volume) is labeled S & M and is their equation $k = 6\ (300/T)^{1.14}$, and represents measurements on polycrystalline rock salt at zero confining pressure. The vertical line is a range of values given by Clark (1966).

possibility exists, of course, that compensating effects have produced coincidental values, and we plan extensive experimental investigation of such a possibility. However, if our thermal conductivity and thermal diffusivity data are accurate, then they give a clear indication that confining pressure in the range 0-50 MPa does not have an important effect on the thermal properties of rock salt. *Supporting the lack of a pressure effect are the* following:

1. *As discussed in the introduction, the model of thermally induced cracking arising from local differences in thermal expansivity, would predict that no fractures would form in pure rock salt during uniform heating.*

2. Thermal expansion does not show a marked pressure effect between 0 and 50 MPa in our experiments and is fairly close to reference values (Fig. 6). While the expansion measurement in our apparatus also lacks an analysis for systematic error, it is simpler and much less vulnerable to systematic problems than is the conductivity measurement. In so-called "hard" rock, such as granite, it is not unusual to observe an increase in the coefficient of thermal expansion with decreasing pressure as local thermal incompatibilities lead to microfracturing and increased crack porosity, hence swelling beyond the intrinsic level (Cooper and Simmons, 1977; Heard, 1980). It can be inferred in the present work that since expansion did not increase with decreasing pressures, heating to 573 K has not produced additional crack porosity. Since crack porosity has not changed with pressure, thermal conductivity and diffusivity would not be expected to change.

3. The data of Yang (1981) are based primarily on single crystal measurements at very high pressures (above 500 MPa) so probably reflect intrinsic values, whereas the data given in Clark [illegible] and by Sweet (1979) are based on measurements made at zero pressure on polycrystalline samples. The intrinsic values agree well with the zero pressure values.

SUMMARY

Preliminary measurements on rock salt between 300 and 573 K under true hydrostatic static confinement from 0 to 50 MPa indicate that pressure is not an important determinant of thermal conductivity, diffusivity, and linear expansivity, ±10% in a coarse-grained rock salt from Avery Island, Louisiana. The possibility of large experimental errors in the conductivity and diffusivity measurements cannot yet be excluded, but the lack of a pressure effect is supported by previous work and by simple models of microcracking in crystalline rocks. The implications for a nuclear waste storage facility in rock salt are positive: except for possible effects of a temperature gradient combined with a nonhydrostatic stress (which has not been explored in this work), the thermal load introduced by the nuclear waste cannot be expected to push thermal conductivity or diffusivity below intrinsic values nor linear expansion above the intrinsic value, regardless of the depth of burial.

It should be emphasized that the same result cannot be expected in most other geological settings being considered as repository sites. The isotropy and monomineralogy of rock salt over extended distances are unique. Most rock types are known to develop microfracture porosity upon heating at low confining pressures. Accompanying thermal effects remain unmeasured.

ACKNOWLEDGMENTS

This work was performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory of the University of California under Contract W-7405-ENG-48. It is a part of the National Waste Terminal Storage Program administered by the Office of Nuclear Waste Isolation, Battelle Memorial Institute.

REFERENCES

Abey, A. E., 1979, Use of a general nonlinear least-squares curve fitting program with nonclosed form relationships, Lawrence Livermore National Laboratory, Livermore, CA. Report UCID-18506.

Clark, S. P., 1966,"Handbook of Physical Constants," Geological Society of America, Memoir 97, New York.

Cooper, H. W., and Simmons, G., 1977, The effect of cracks on the thermal expansion of rocks, Earth Planet Sci. Lett., 36: 404-412.

Hasselman, D. P. H., Effect of micro-cracking on thermal conductivity: analysis and experiment, this volume.

Heard, H. C., 1980, Thermal expansion and inferred permeability of Climax Stock quartz monzonite to 300°C and 27.6 MPa, Int. J. Rock Mech. Min. Sci., 17:289.

Kaufmann, D. W., ed., 1960, "Sodium Chloride," Reinhold Publishing Corporation, New York, p. 367.

Sweet, J. N., 1979, Pressure effects on thermal conductivity and expansion of geologic materials, Sandia Laboratories Energy Report SAND78-1991, Sandia Laboratories, Albuquerque, NM.

Sweet, J. N., and J. E. McCreight, Thermal conductivity of rock salt and other geologic materials from the site of the proposed waste isolation pilot plant.

Walsh, J. B., and W. F. Brace, 1966, Elasticity of rock: a review of some recent theoretical studies, Rock Mech. Engr. Geology, IV/4:283.

Walsh, J. B., and E. R. Decker, 1966, Effect of pressure and saturating fluid on the thermal conductivity of compact rock, J. Geophys. Res., 71:3053.

Yang, J. M., 1981, Thermophysical properties, in: "Handbook of Rock Salt Properties Data," L. H. Gevantman, ed., Chpt. 4, National Bureau of Standards.