MATERIAL PROPERTIES OF ELEANA ARGILLITE--EXTRAPOLATION
TO OTHER ARGILLACEOUS ROCKS, AND IMPLICATIONS FOR WASTE MANAGEMENT*

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ABSTRACT

Results of a near-surface heater test in the Eleana argillite suggest the possibility that the high-temperature ($> 100^{\circ}\text{C}$) thermo-mechanical response of argillite to waste emplacement may be dominated by behavior of expandable clays. Enough expandable clay is probably present in most argillaceous rocks to cause a similar response. In situ thermal conductivities may be markedly reduced by even small amounts of clay contraction which results in opening of pre-existing joints. A simple model predicts that such behavior may continue to operate to considerable depths, though several factors affecting determination of this depth remain poorly defined at present.

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Introduction

Results of a full-scale (3.5 kW) near-surface heater experiment recently completed in argillite of the Eleana Formation on the U.S. Department of Energy Nevada Test Site indicate that the thermo-mechanical response of many argillaceous rocks to the emplacement of heat-producing nuclear wastes may be dominated by effects resulting from the contraction of expandable clays. This conclusion is applicable only if power densities are sufficient to cause boiling of water within the formation or significant rock dehydration at lower temperatures, and is based on a consistent correlation between lab-measured thermal expansion of argillite and field observations related to the Eleana heater test. These observations indicate:

- 1) opening of joints in the heater hole wall, as a result of volumetric contractions;
- 2) apparent decrease of in situ thermal conductivity of the argillite below reported values for temperatures above approximately 100°C;
- 3) marked increase in argillite permeability inside the 100°C isotherm, from the millidarcy to darcy range; and
- 4) presence of a rubble zone near the heater, encountered in posttest drillback and coring operations.

The objectives of this paper are to: 1) examine the amounts of expansion/contraction that might be expected to occur for a range of argillaceous rocks on dewatering of the expandable clays; 2) estimate some of the effects of this contraction on in situ thermal conductivity; and 3) gain some feel for the extent to which tensional behavior may occur as a result of experimentation or waste emplacement in argillaceous rocks at depth.

Thermal Expansion/Contraction of Argillaceous Rocks

Thermal expansion of silicate rocks is complex. It may be strongly affected by variables such as bulk confining pressure, which affect the state of the matrix, porosity, and microcracks present,¹ and also by the local fluid or water pressure at the time of heating. The temperatures at which the expandable clays within argillaceous rocks dehydrate and contract are strongly dependent upon fluid pressure. Volume and length changes associated with this dehydration depend upon the amount of expandable clay present.

Measurements of the thermal expansion of argillaceous rocks are extremely limited in number. Results of a series of ambient-pressure measurements on Eleona argillite are shown in Figure 1. Sample SI-72 is from the central heater hole of the full-scale heater test, 1 m above the heater center plane. "Natural state" samples which were waxed and wrapped until prepared for measurement, are indicated by solid lines in Figure 1 and show variable linear contraction beginning soon after heating. This contraction reflects the de-watering and contraction of expandable clays, which begin at very low temperatures in these tests conducted at ambient pressure. After reaching maximum contraction at or near 100°C, the samples then expand continuously to 500°C, with a linear expansion coefficient (α_L) of between 8 and 15 x 10⁻⁶ °C⁻¹.

Anisotropy of expansion varies markedly in the Eleona. Sample Uel7e-2200 is not measurably anisotropic: three mutually perpendicular blanks of this sample have the same linear expansion coefficient within $\pm 1.0 \times 10^{-6}$ °C⁻¹, the approximate limit of accuracy of these measurements. Much of the Eleona appears to be similarly massive,

Figure 1. Ambient-pressure linear thermal expansion of Eleena argillite to 500°C. Solid lines="natural state" samples, waxed and wrapped until prepared for measurement; Dashed lines= air-dried samples; Heavy dashed line = expansion behavior assumed in modeling of Eleena heater test.

with macroscopically indistinguishable bedding. Macroscopically layered samples, however, generally demonstrate anisotropic expansion. For example, sample Uel7e-993 contracts $1.2 \pm 0.2\%$ parallel to layering upon long-term heating at 110°C , and $1.9 \pm 0.2\%$ perpendicular to layering. These values correspond to α_L values of -130 and $-210 \times 10^{-6} \text{ }^{\circ}\text{C}^{-1}$, respectively, between ambient and 110°C .

Sample state also plays a major role in results of expansion measurements of argillaceous rocks. For example, Uel7e-370, which is strongly layered and was air-dried for approximately six months before measurement, does not show contraction at or below 100°C , but does display marked anisotropy above 400°C , presumably due to structural breakdown of constituent clays. Above 400°C , the blank of sample 370 cut perpendicular to layering contracts, while that cut parallel to layering continues to expand. Also shown in Figure 1 is the expansion behavior assumed for purposes of mechanical modeling of the full-scale heater test in the Elcena Formation. Note that it was assumed that dehydration of the in situ argillite did not begin until 75°C .

Mineralogical analyses of the Elcena argillite show a broad variation in assemblage. The general assemblage is: quartz + "illite" $\pm$ kaolinite $\pm$ chamosite $\pm$ pyrophyllite $\pm$ calcite $\pm$ vermiculite $\pm$ siderite $\pm$ calcite $\pm$ ferroaluminosilicate $\pm$ pyrite. Quartz content ranges approximately from 15 to 50 weight percent. From a thermal expansion point of view, the most important phase in argillite would appear to be the "illite", which contains appreciable expandable interlayers of vermiculite or montmorillonite. Bulk X-ray analysis of powdered samples indicates that the basal spacing of

the "natural state illite" is 10.5 to 10.8 Å at ambient temperature, decreasing to 10.0 - 10.2 Å after heating to 400°C. Based on the interpretive curves of Jonas and Brown,² this behavior indicates that the "illite" contains up to about 10% interlayering of chlorite, and that 60 - 80% of the "illite" is indeed illite (nonexpandable), while the rest is an expandable phase (montmorillonite or vermiculite).

While there is generally less expandable clay in Paleozoic shales than in younger material, it would appear that enough should be present in most older rocks,^{3,4} such as Eleana argillite (Mississippian age), to result in there being no net expansion upon heating to 100°C or the boiling point of water. In fact, there may be a net contraction of many rocks. Assuming that the volumetric expansion coefficient (α_v) of an average argillaceous rock at temperatures below boiling is $36 \times 10^{-6} \text{ } ^\circ\text{C}^{-1}$, and that the boiling point is 100°C, then the net volumetric expansion between ambient temperature and the beginning of boiling will be approximately 0.00% cm³/cm³. If the basal spacing of the expandable clays (montmorillonite or vermiculite) present in this rock as discrete phases or as interlayers with a nonexpanding phase is assumed to contract from 12.5 to 10 Å upon dehydration near 100°C, then only 1.5 volume percent of the rock need be expandable clay in order for the rock to experience no net expansion to that temperature. Higher content and larger initial basal spacing of the expandable phase(s) would lead to absolute contraction. Retention of partially expanded basal spacings to higher temperatures would reduce the amount of contraction.

Considering an "average" shale,⁵ which contains approximately 70% clay mineral, only 2.5% of the total clay content must be expandable to result in no net expansion at 100°C under these assumptions. If "illite" makes up 25% of the total clay assemblage, a reasonable lower limit,⁴ then 10% interlayering of expandable phase in the "illite" is sufficient. If "illite" comprises 80% of the clay assemblage, which is quite common in lower Paleozoic shales, then only 3% interlayering is required.

It thus appears as if virtually any argillaceous rock, even a Paleozoic shale generally considered to contain "no" expandable clay, may be subject to the same sort of contraction behavior as seen in the Eleena near-surface heater test. This is based on the fact that the smallest amount of expandable-phase interlayering that can be detected by X-ray analysis is approximately 10%,^{6,7} and that at least slight interlayering is generally reported in even deep-seated shales. For example, assuming that the clay minerals are randomly oriented, illitic shales of the Oligocene Erio Formation, encountered in deep wells along the Gulf Coast of the United States, should undergo $0.4 \pm 0.1\%$ linear contraction, based on their mineralogical composition and reported uncertainties in extent of interlayering of expanded phases.⁸ Representative Pierre shale containing approximately 75% total clay, of which 75% is mixed-layer illite containing 60% Montmorillonite interlayers,⁹ should undergo $2.2 \pm 0.3\%$ linear contraction after dehydration of expandable phases.

Effects of Contraction on In Situ Conductivity

Contraction of argillaceous rocks near the boiling point of water

will have an appreciable effect on in situ thermal conductivities only if the contraction of heated domains is not compensated by general infilling from surrounding areas. In the Eleena heater test, contraction of clays within discrete joint-bounded blocks appears to have resulted in simple opening of pre-existing joints, with perhaps some additional breakage as well. In the following discussion, similar behavior is assumed to be realistic for a range of argillaceous rocks, and thermal effects resulting from the opening of pre-existing joints are briefly discussed. A simple one-dimensional analysis of changes in thermal conductivity due to the presence of planar joints perpendicular to the direction of heat flow is given.

Little information is available concerning the matrix conductivity of argillaceous rocks at or above 1000°C. Some of the available information is summarized in Table 1. As shown, the reported range of conductivities at elevated temperature is from 0.3 to 2.1 W/m°C.

If it is assumed that heat flow within a rock mass is perpendicular to planar joints, that the joints are small enough in aperture to not allow convection, and that thermal radiation is negligible, then the effective conductivity of the jointed mass can be approximated by use of the formula for heat flow perpendicular to the layers of a laminated structure:

$$K = \frac{1}{\frac{X_1}{K_1} + \frac{X_2}{K_2}} \quad 1.$$

where K_1 and K_2 are rock matrix conductivity and saturated steam

Table 1
Matrix Thermal Conductivity of Argillaceous Rocks at
or Above 100°C

<u>Rock Types</u>	<u>k (W/m°C)</u>	<u>Reference</u>
Boom Clay (Belgium)	0.73	10
Slate (Perpendicular to bedding)	1.70	11
Eleona Argillite		
(Parallel to Layering)	1.10	12
(Perpendicular to Layering)	1.14	
Conasauga Shale (calcareous)	0.70-1.1	13, 14

conductivity at 100°C, respectively and λ_1 and λ_2 are the fractional lengths of rock matrix and joint per unit length of rock mass. Thermal conductivities calculated as a function of percent linear contraction and matrix conductivity at 100°C, by means of equation 1, are shown in Figure 2. The specific matrix conductivities indicated are 2.3 W/m°C for the Boom clay (Belgium) and 1.75 for Eleana argillite parallel to layering. Curves are also drawn for initial matrix conductivities (100°C) of 0.75, 1.1 and 2.5 W/m°C. The solid curves in Figure 2 indicate that absolute decreases in conductivity are greatest at constant percent joint opening (or linear contraction) for rocks with the highest matrix conductivities. As indicated by curves a and b in Figure 2, greater contraction is also required to produce a constant relative decrease in conductivity, as matrix conductivity decreases.

The effects of even small amounts of contraction could be significant. For example, if it is assumed that shale from the Erie Formation had a matrix conductivity of 2.0 W/m°C at 100°C, the estimated amount of contraction of this shale could reduce this to near 1.5 W/m°C, a 25% reduction. In the case of Eleana argillite, 13 contraction parallel to layering could reduce in situ conductivity from 1.75 to about 1.65 W/m°C, a 40% reduction. Contraction perpendicular to layering in the Eleana could reduce the effective conductivity from 1.5 to 0.75 W/m°C, a 50% reduction. If contraction and joint opening are not considered, the ratio of conductivities perpendicular and parallel to layering in the Eleana argillite (1.50/1.75) is 0.86 at 100°C. If contraction is considered and it is assumed that joints perpendicular to bedding are also perpendicular to heat flow in the layering, the ratio decreases to 0.71. Decreased sensitivity of in situ conductivity for rocks with a relatively low matrix conductivity to uncertainties caused by thermal contraction and jointing may partially offset advantages of waste emplacement in higher-conductivity media.

Additional factors would appear to add uncertainty to the estimates of in situ conductivity based solely on matrix conductivity and mineralogy. For example, heat flow in real or simulated waste emplacement, such as the Eleana heater test, is not constrained to be perpendicular to any joint sets, nor are joint sets necessarily continuous. Contraction of clays appears to result predominately in opening of the pre-existing joint sets, rather than in generation of new joints. This fact obviously requires that future experimentation in argillaceous rocks include careful characterization of the pre-existing joint sets and consideration of the impacts of their possible effects on heat flow in different directions relative to canister orientation.

In the case of the Eleana argillite, joint structure frequency and orientation taken from vertical drill holes indicate that some 70% of all fractures in the argillite are parallel to bedding.^{15,16} At most depth intervals in these holes, however, there is a distinct subgroup of joints that is vertical or nearly so. Similar predominance of bedding plane joints appears to be the case in many argillaceous rocks.¹⁷ Joint (bedding plane parting) frequencies of the order of 30/m are not uncommon parallel to layering. Steeply dipping joints, which tend to occur in two distinct sets, often occur at spacings of 1 m or more. Frequently, only one vertical set is well-developed, regular, and relatively continuous, and the other is much less prominent. One possible result of this is that considerable directional anisotropy may be superimposed on the thermal response parallel to layering in and above a repository in argillaceous

Figure 2. Approximate rock mass conductivity at 100°C as a function of matrix conductivity and extent of joint opening. Curves a and b indicate the amount of opening required for 10 and 20% reduction, respectively, of rock mass conductivity. Indicated ranges of conductivity of specific rocks are meant to indicate nothing about their conductivity.

rocks. For jointed rock masses as treated thus far, the calculated ratio of conductivity perpendicular and parallel to layering in the Eleana is 0.71. If, however, heat flow parallel to layering is also parallel to the vertical joint set, the ratio decreases to 0.43. This directional variation would cause the in situ temperature distribution to be a three-dimensional function of heat flow, clay content, and joint orientation.

Contraction and Tensile Joint Opening at Depth

While opening of joints due to volumetric contraction appears to have taken place in the Eleana heater test, the emplacement depths to which similar behavior would be observed in the Eleana, or any other argillaceous rock type, are unclear at present. A simple, one-dimensional analysis, in part compared with detailed thermomechanical modeling of the Eleana heater test, indicates that such opening is not likely to be solely a near-surface phenomenon. In this analysis, the maximum tensile stress resulting from heating is taken to equal the product of the Young's modulus and maximum linear contraction of a given rock type, and is compared with assumed hydrostatic pressures as a function of depth. It is further assumed that tensional joint opening will occur if the tensional stress exceeds the sum of the hydrostatic stress plus in situ tensile strength of the rock. The resulting failure criterion, for an assumed in situ tensile strength of 3.5 MPa (500 psi), is shown in Figure 3 and compared with the results of thermomechanical modeling of a configuration the same as the Eleana heater test, at a series of depths. In this figure, the full and open circles represent results of two-dimensional axisymmetric thermomechanical modeling of the heater test.¹⁸ The

● Results of modeling
with AGINA, mod 5

Figure 3. Maximum possible thermally induced tensile stress versus depth. Above solid line, joints are predicted to open, due to volumetric contraction. Below it they are not.

calculations were made with the ADINA code¹⁹, which includes a tension cut-off. For these calculations this was set at 0.1 MPa. The simplified assumptions predict that, for an assumed Young's modulus and maximum linear contraction of the Eleana of 7.20 GPa (1×10^6 psi) and 1.0 percent, respectively, the maximum possible tensile stress should be 69 MPa (1×10^4 psi), and some tensional opening should occur for emplacement at depths up to 2900 m (9500 feet). Variations in clay content will change the depths to which opening persists for a rock of constant Young's modulus. For example, a rock which has a Young's modulus of 6.90 GPa, but which contracts only 0.1% upon dehydration, should exhibit a maximum tensile stress of only 6.90 MPa (1×10^3 psi), and opening of joints to a depth of only about 596 m. Calculation of variation in the depth to which tensile openings persist as a function of clay content are relatively straightforward.

Many factors, however, will influence the in situ Young's modulus of argillaceous rock and, in turn, cause changes in the depth boundary between open and closed joints shown in Figure 1. Intact samples of most shales have unconfined Young's modulus in the range of 0-50 GPa.²⁰ The lab-measured modulus of the Eleana, 7.2 GPa, falls near the low end of this range and is also low compared to a "typical silicic shale"; see Table II. However, the modulus of the Eleana increases rapidly with increasing confining pressure, and for pressures greater than about 20 MPa, is constant at about 16 MPa. The phenomenon of a constant modulus above a certain confining pressure has also been observed in other argillaceous rocks.^{21,22} It should be pointed out that the depth corresponding to the pressure at which the modulus of the matrix becomes constant (~ 0.9 km) is typical of depths dis-

Table II

Comparison of Eleana Argillite to "Typical Illite Shale;" Shale Data from Reference 17.

<u>Unconfined</u>	<u>Eleana</u>	<u>Illite Shale</u>
Young's Modulus (GPa)	3.74	11.03
Poisson's Ratio	0.30	0.15
Compressive Strength (MPa)	4.75	69.0
Cohesion (MPa) (Intersection of Mohr-Coulomb Failure Envelope with Shear Stress Axis.)	0.8	41.0
<u>Confined</u>		
Bulk Modulus (GPa)	14.0	5.20
Angle of Internal Friction (deg)	35	26

cussed for potential repositories and, hence, that a pressure-dependent modulus may be operative at the disposal horizon. A pressure-dependent modulus obviously should be used in mechanical modeling of overlying layers.

Moisture content has recently been shown to be a very important factor in altering the elastic compliance matrices for three argillaceous rocks from coal mines in the U.S.²¹ The tests were run in unconfined compression at room temperature. When the relative humidity was varied from 0 to 100%, Young's modulus measured perpendicular to bedding was reduced by at least 50% for each of the three shales. Some of the compliances were even more strongly affected. The implication of this sensitivity is that the moduli of argillaceous rock can be time-varying due to changing water contents, even at a given P and T.

In situ stress, both pre-existing and resulting from repository excavation and waste emplacement, may have a strong influence on thermal conductivity of argillaceous rocks, through its effect on dilatancy (microcrack opening) and joint dilatancy. Microcrack dilatancy depends on both the orientation and magnitude of the deviatoric part of the stress tensor (e.g., reference 24). Joint dilatancy, on the other hand, results from the riding of adjacent blocks over asperities on the joint surface between them, and is more closely related to the amount of shear displacement.²⁵

Our findings to date with regard to stress-volume relationships for Eleana argillite are summarized in Figure 4. The specimens tested were right circular cylinders 5 cm in diameter X 10 cm long and, though coherent, contained macroscopic joints at a natural joint spacing of about 2.5 cm. Several distinct joint orientations were evident in each sample, giving the specimens a blocky appearance. In Figure 4,

Figure 4. Mean stress versus volume for ambient-temperature testing of Eleana argillite. Compaction is considered to be positive.

the vertical axis is mean stress ($\sigma_m = (\sigma_1 + \sigma_2 + \sigma_3)/3$) and the horizontal axis is volumetric strain, ϵ_v , in percent. Compaction is considered to be positive. The thick line is the common hydrostat for the four tests shown. The tests were run in confined compression, so that the first phase of each test is hydrostatic loading up to the predetermined confining pressure (0, 20, and 33 MPa), marked by triangles in the figure. The thin lines, one for each test, show the $\sigma_m - \epsilon_v$ relations for the deviatoric stress phase of each test. Positive slopes represent compaction, and negative slopes represent expansion or dilatancy. Curve I represents results of an unconfined compression test and shows that initial axial compression causes a decrease in volume, as a result of crack closure. At about 50% of the specimen strength, dilatancy sets in and the volume increases continuously until the ultimate strength is reached. Test IV, which was run at a confining pressure of 33 MPa, shows that the specimen compacts during hydrostatic loading, and continues to compact during deviatoric loading. Transitional behavior is exhibited by tests II and III, run at a confining pressure of 20 MPa. Test II showed slight compaction during early deviatoric loading, followed by expansion, in contrast Specimen III showed continuous compaction throughout.

Apparently 20 MPa (~ 3000 psi) represents a critical confining pressure above which specimens of Eleana argillite compact in response to deviatoric loading at room temperature, and below which they dilate. If a repository in Eleana argillite were less than 900 m (3000 feet) deep confining pressures would be less than ~ 20 MPa, and dilatancy could be a predominant response to deviatoric stresses. Also, near-field stresses resulting from excavation and waste emplacement should tend to be more deviatoric than those of the far-field. Hence, dila-

tancy problems should be enhanced in the near-field, i.e., in that area that is most likely to undergo sufficient heating to cause contraction of expandable clays.

At least two major factors that might affect the response of argillaceous rock to waste emplacement remain poorly defined. Little work has been done on the mechanical properties of argillaceous rock at elevated temperature. Although it has been shown²² that the strength of some shales decreases markedly with increased temperature, the temperature dependence of the modulus is not documented. For many other rock types, in fact, increasing confining pressure seems to have a stronger effect on elastic moduli than increasing temperature. The effect of time, as examined in creep tests or in constant deformation rate tests of argillaceous rocks, is also not well documented. One recent study²⁵ does provide a limited amount of creep data on the Ophir Shale. Although some of the results of this study are difficult to interpret, they do indicate that steady-state creep rates at very low deviatoric stresses are on the order of 10^{-9} s^{-1} . One observation of this study is that, when the shale is subjected to a hydrostatic pressure step-function history, the resulting volume-time curve looks just like an ordinary creep curve, i.e., volume compaction has a time-dependent, viscoelastic nature. This is an important result because most creep testing programs have ignored the fact that the general viscoelastic constitutive relation of rocks includes a time-dependent bulk modulus, as well as the time-dependent Young's modulus.

Conclusions

Though the treatment presented here is simple and subject to many uncertainties and uncontrolled or poorly defined variables, three main

conclusions appear to be justified by available data:

1) Expandable clays, such as montmorillonite or vermiculite, may play a strong role in the thermomechanical response of argillaceous rocks to the emplacement of nuclear wastes. This is especially true if the rocks are heated above the boiling point of water. Expandable phases may occur as discrete phases, or as interlayers in mixed-layer clays, even at small degrees of interlayering commonly not detectable by means of X-ray diffraction.

2) Dewatering and contraction of expandable clays, if accommodated by opening of the pre-existing joints inherent to argillaceous rocks, could cause a reduction of in situ thermal conductivity. Both absolute and relative decreases would be greater for rocks with higher matrix conductivity. Variations in directional heat flow relative to pre-existing joints could result in a highly anisotropic thermal distribution. Decreases of in situ conductivity as a result of heating due to waste emplacement could occur for disposal at considerable depths. At constant Young's modulus, the depth to which joint opening would continue to occur decreases with decreasing content of expandable clay.

3) In addition to the effects of variable Young's modulus and clay content, variables such as confining pressure, non-hydrostatic stress, rock moisture content, time, temperature, and deformation rate will influence the presence or absence of dilatant behavior at depth. These factors are poorly defined at present.

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