Structural Response of Rich Green River Oil Shales to Heat and Stress and Its Relationship to Induced Permeability

PETER R. TISOT and HAROLD W. SOHNS
Laramie Energy Research Center, Bureau of Mines, U. S. Department of the Interior, Laramie, Wy. 82070

Structural response and/or deformation of three rich oil shales to heat and compressive stress was determined. Specimens consisted of small cores and of small columns of fragments from oil shales assaying 34.5, 45.5, and 63.5 gallons of oil per ton. Effect of structural deformation on induced permeability in the fragmented specimens was determined. Stress-strain-time-temperature relationships of these oil shales provided information regarding their yield temperature, yield stress, rate of compressive strain, and loss of mechanical strength as they were heated from ambient to subretorting temperatures—that is, below those required for rapid pyrolysis of the organic matter to produce oil. Structural response of some cores was also observed as they were heated in a stress-free environment. Results provide guidelines to assess and evaluate some problems that may be associated with recovery of shale oil by underground retorting methods.

GREEN RIVER oil shale is a highly consolidated rock composed of a complex mixture of organic and inorganic constituents in variable proportions. Considerable research has been conducted on aboveground methods of retorting, and many such systems have been developed, some to commercial or semicommercial scale (*1, 2, 6, 7, 11*). A second approach for shale oil recovery, which in recent years has been given increasing consideration, is underground processing. The objectives of this approach are threefold: to utilize deep deposits; to eliminate mining, transporting, and crushing of the raw oil shale; and to eliminate disposing of the spent rock.

Because the rock formation is impervious, present oil shale technology requires that permeability be induced within the process area. One possible method to induce permeability includes conventional hydraulic fracturing to open up fractures through which the heat transfer agent and products are passed between injection and producing wells. In a second method, a nuclear explosion is used to create large columnar masses of broken rock or rubble. Predictions have been made of the geometric configuration, size, and quantity of fragmented rock that may be expected from a nuclear explosion in oil shale (*3, 4, 5, 8*). The bulk porosity of a rubble system has been calculated to be 25 to 30% (*4*); therefore, induced permeability should be high.

Many fundamental engineering data need to be developed to understand and assess the feasibility of designs for underground retorting using either technique to induce permeability. Many problems need to be considered for which little or no previous experience exists. One such problem, the basis of this paper, is the support capability of the column of rubble as it is being processed. Raw oil shale has high compressive strength (*12*)—many times that required to withstand the stresses present in the column of rubble. However, very little is known regarding the structural response of oil shale, particularly oil shales of high organic content, as they are heated under compressive stress. Knowledge of whether or not extensive structural deformation would occur and the effect of such deformation on the induced permeability is of prime importance for intelligent engineering design.

An investigation was conducted, utilizing small specimens, to evaluate stress-strain-time-temperature relationships of three rich oil shales under compressive stresses calculated to exist in the column of rubble and to study the effect of deformation on induced permeability. The primary objective of this research was to obtain information regarding structural behavior of rich oil shales as they were heated from ambient to subretorting temperatures—that is, below those required for rapid conversion of the organic matter to oil. In most tests the specimens were heated to 725° F., whereas rapid pyrolysis of the organic matter occurs from 825° to 900° F. Such information is useful to predict structural response of rich oil shales ahead of the retorting zone.

A probable area for a nuclear explosion is the lower oil shale zone in the Colorado Piceance Creek Basin; however, samples from this zone were not available for laboratory study. Therefore, the rich oil shale samples were selected from the Bureau of Mines experimental mine near Rifle, Colo. The oil shales' Fischer assays, to the nearest ½ gallon, were 34.5, 45.5, and 63.5 gallons of oil per ton. The samples from the experimental mine were considered to be comparable, grade for grade, to those that occur in the lower zone of the basin and, therefore, the experimental results would be applicable to both oil shales.

A histogram showing weight per cent organic matter and oil yield from two 100-foot intervals in the lower oil shale zone of the Colorado Piceance Creek Basin (*10*) is presented in Figure 1. About 66% of the oil shale within these two intervals is of the same grade as that used in this investigation.

EXPERIMENTAL

Specimen Preparation. From each of the oil shales (34.5, 45.5, and 63.5 gallons per ton), a sample 2 inches thick and approximately 18 inches square was cut with its long axis parallel to the bedding plane. Care was used in the

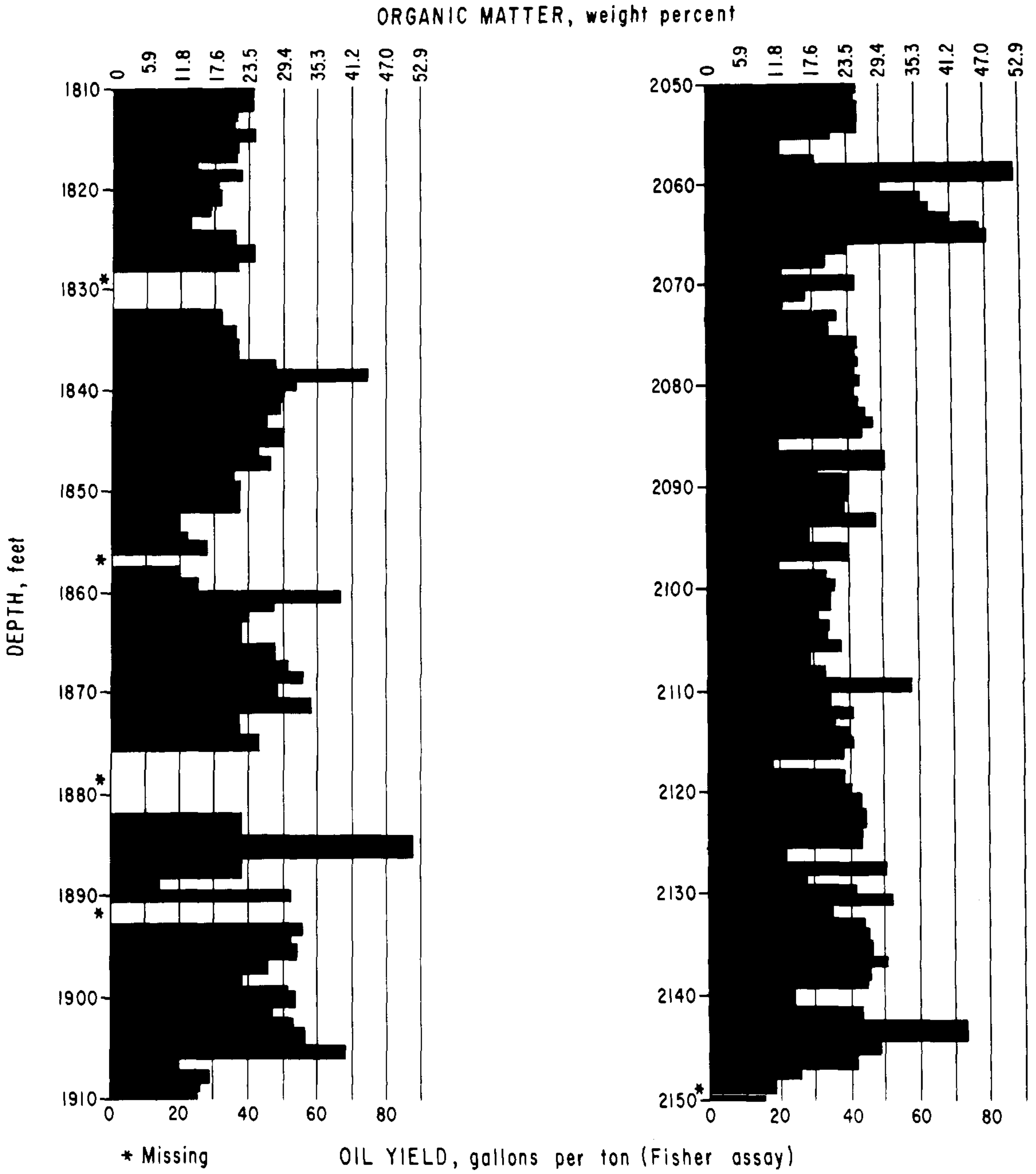

Figure 1. Histogram of organic matter and oil yield of typical lower oil shale zone in Colorado Piceance Creek Basin

selection to ensure that each sample appeared homogeneous with respect to distribution of the organic matter and mineral constituents and that it contained no fractures due to relaxation of residual stresses in the rock. From these samples, specimens were prepared as cores, ¾ inch in diameter and 1½ inches long, cut perpendicular to the oil shales' bedding plane and as small fragments, $^{3}/_{16}$ to ⅜ inch long, along their major axes.

Compressive Strength. Compressive strength of the three rich oil shales was determined with a hydraulic compression tester. Cores, ¾ inch in diameter and 1½ inches long, were carefully ground to assure that the flat surface on each end was smooth and normal to the long axis. Four cores were tested for each oil shale grade. The specimens were uniaxially stressed, perpendicular to the bedding plane, at a rate of 1200 to 1500 p.s.i. per minute to structural failure.

Stress-Strain Apparatus. A stress-strain microunit (Figure 2), equipped with an electric heater, was designed and fabricated in the laboratory such that compressive stress, temperature, rate of heating, and gaseous environment could be controlled. The apparatus was insulated. Part of the insulation included an evacuated double-walled cylinder placed between the heating element and the outer shell of the unit. Compressive stress, measured with a proving ring, was transmitted to the specimens through a gas-activated piston. Compressive strain was measured with a dial gage, sensitive to 0.0025 mm. A sensitive rotameter measured the incoming gas flow to the fragments. The gas escaped at the upper end of the specimen.

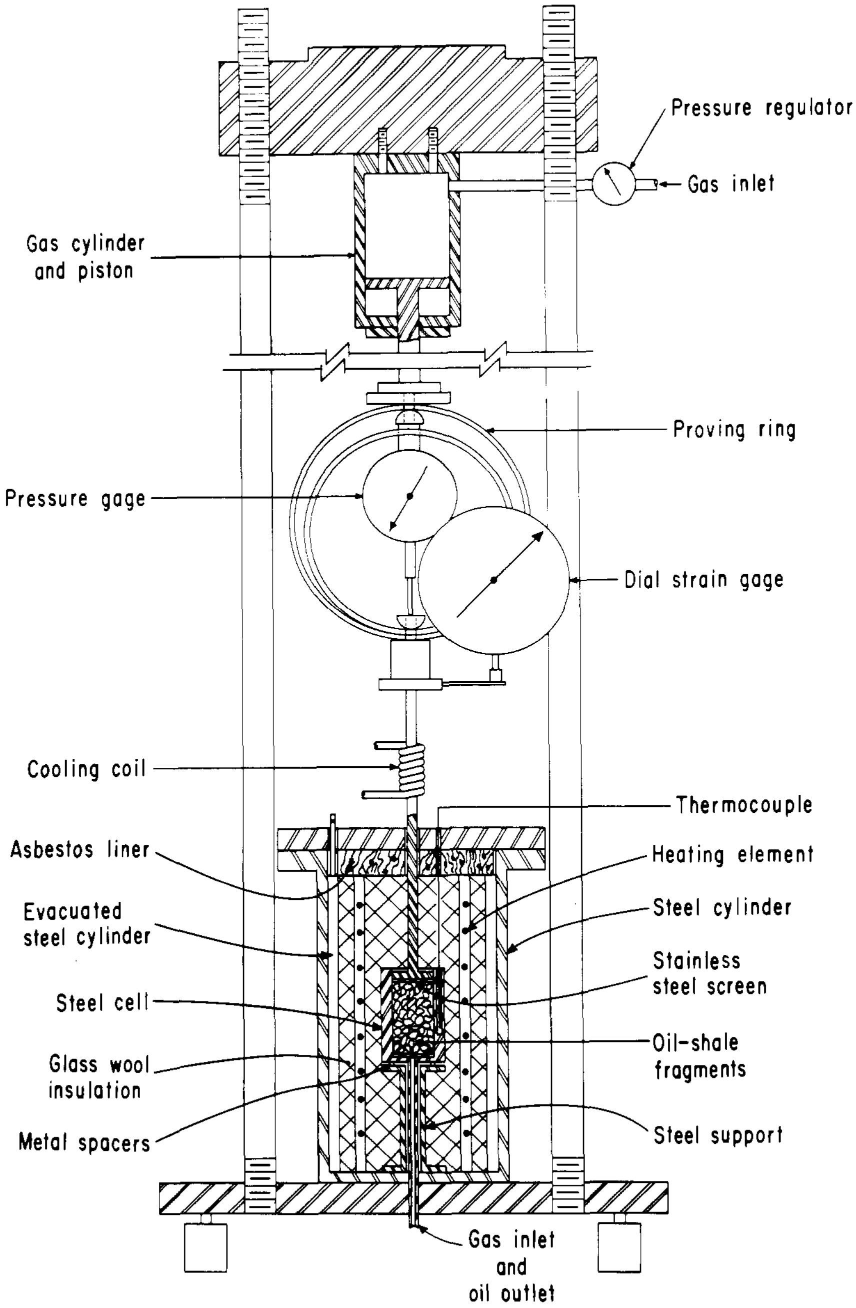

Figure 2. Schematic diagram of apparatus for testing structural response of rich oil shales under heat and stress

Operating Conditions. Many combinations of compressive stress, heating rate, and temperature may be selected for investigating the structural response of rich oil shales to heat and stress. This study was primarily concerned with the structural response of these oil shales at temperatures below those required for rapid pyrolysis of the organic matter and under compressive stresses expected to exist within the rubble column. The specimens were heated from ambient temperature to 725° or to 825° F. in an inert atmosphere at a heating rate of 2° F. per minute.

Structural response of some cores to heat was observed in a stress-free environment—that is, with no compressive stress imposed on any of their surfaces. All other specimens (both cores and fragments), prior to and during heating, were stressed under constant load of 80, 200, and 325 p.s.i. These stress levels were calculated to approximate those at the 100-, 250-, and 400-foot depths in a rubble column. Permeability measurements in most tests were taken with a pressure differential across the column of 3 p.s.i. Some permeabilities were measured with a pressure differential of 14.7 p.s.i.

Treatment of Stress-Free Cores. Cores, $\frac{3}{4}$ inch in diameter and $1\frac{1}{2}$ inches long, were heated in a stress-free environment to observe the effect of heat on deformation due to expansion along both the major and minor axes. Some cores were heated with no restraint imposed on them during the entire test. Other cores were first placed upright in a small vise-like frame which restrained movement only along the major axis. Although stress-free initially, these cores developed stress in their attempt to swell along the major axis as they were heated. Specimens were placed in an electric muffle and heated in an inert atmosphere to either 725° or 825° F. They were removed from the electric muffle after the major portion of the organic matter was pyrolyzed.

Treatment of Uniaxially Stressed Cores. Cores from the three oil shales were individually tested to observe their structural response to heat and compressive stress. The specimens were uniaxially stressed to constant load (80, 200, or 325 p.s.i.), and then heated in an inert atmosphere from ambient temperature to 725° F. Some cores were maintained at 725° F. to determine the time required for structural failure to occur at this temperature, whereas in other cores heating was continued beyond 725° F. to determine the maximum temperature that could be reached before structural failure.

Treatment of Confined Oil Shale Fragments. Fragments, $\frac{3}{16}$ to $\frac{3}{8}$ inch, were packed at random in a small steel cell, $\frac{3}{4}$ inch in diameter and $2\frac{1}{8}$ inches deep. Stainless steel screens were placed at the top and bottom of the fragments to separate them from direct contact with the base of the steel cell and the piston at the top of the column of fragments. The initial porosity for each fragmented specimen was determined from the volume of the cell (bulk volume) and the weight-density ratio (material volume) of the specimen. The charged cell, containing the 2-inch column of fragments, was placed in the microunit, stressed under constant load to 80, 200, or 325 p.s.i., and then heated to 725° or 825° F. Nitrogen was passed through the column at a rate of 0.04 to 0.05 cu. inch per sq. inch of surface per minute and maintained at this rate until the test was terminated or until flow through the column reduced to zero. In tests where the latter occurred, a pressure differential across the column was imposed of either 3 or 14.7 p.s.i. until the test was terminated.

RESULTS AND DISCUSSION

Compressive Strength of Raw Oil Shales. The compressive strengths of four cores from each of the three oil shales were measured. The average compressive strengths of the

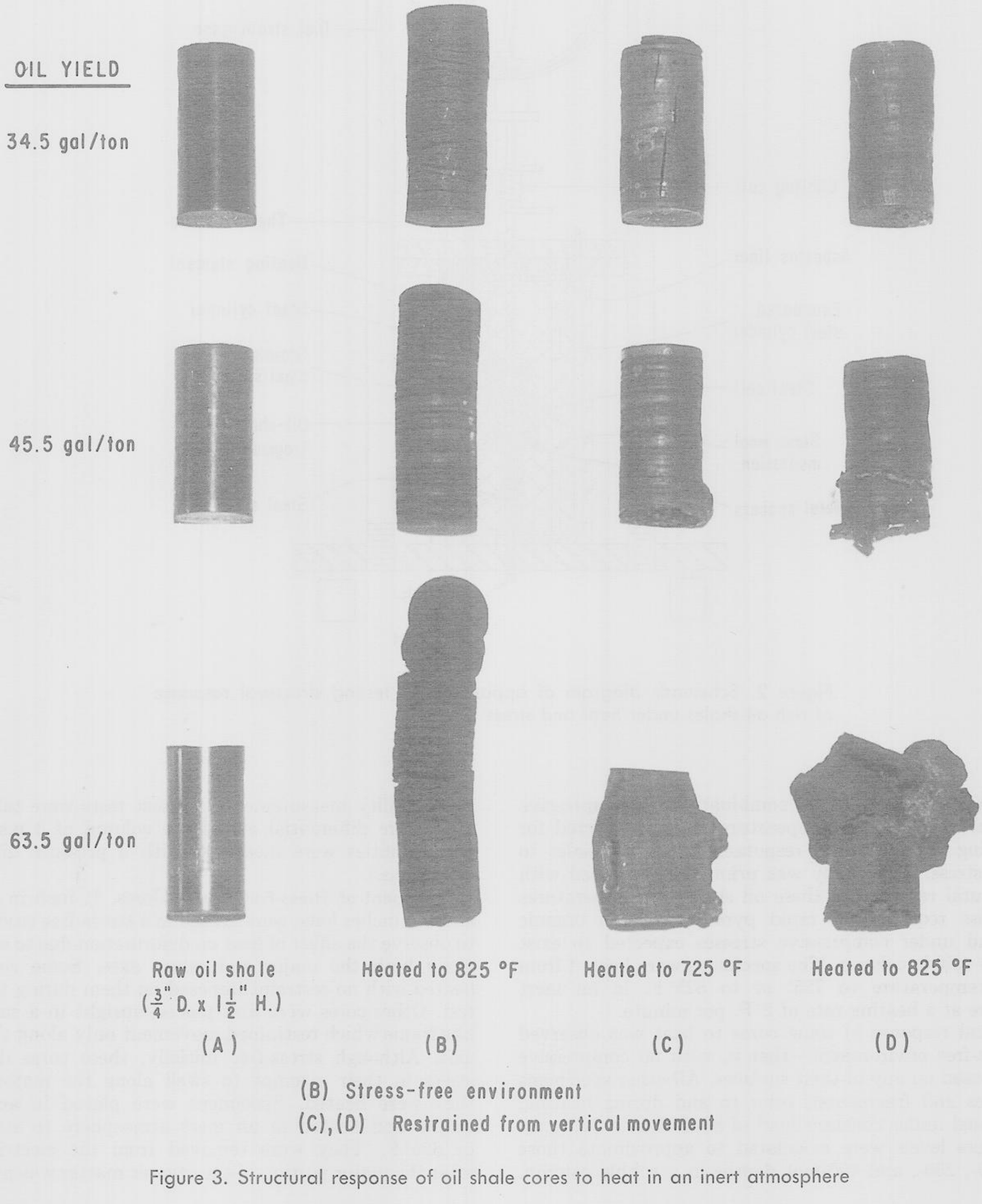

Figure 3. Structural response of oil shale cores to heat in an inert atmosphere

34.5-, 45.5-, and 63.5-gallon-per-ton oil shales were, to the nearest 50 p.s.i., 11,300 10,000, and 9850 p.s.i., respectively. These values, about twice those for high strength concrete, far exceed the support capability required to withstand the stresses encountered in a column of rubble. Compressive strengths taken either perpendicular or parallel to the oil shale's bedding plane do not differ greatly (*12*).

As each specimen was subjected to increasing compressive stress, strain was apparent in all three oil shales. The highest compressive strain, prior to structural failure, was in the richest oil shale, about 14.5% of its initial height. Structural failure was attributed to conical-type failure. In specimens where the load was removed immediately at the point of maximum stress, the cores exhibited elastic rebound, which indicated that under stress they absorbed energy. These cores regained essentially 100% of their initial height.

Response of Stress-Free Cores to Heat. Structural deformation, either constrictive or expansive, is not easily induced in the initial rock by mechanical means. Expansive deformation, however, will occur in stress-free rich oil shales simply by the application of heat. This is accompanied by pronounced deterioration in the mechanical structure of the specimens. Figure 3,*B*, clearly depicts the extensive structural breakdown in a stress-free environment. Deformation occurred preferentially perpendicular to the bedding plane. Many of the fractures shown in the photograph began between 350° and 450° F.

Figure 3,*C*, and *D*, illustrates structural response when movement perpendicular to the bedding plane was restricted. These specimens, although stress-free initially, were subjected to increasing stress as heating was continued. At some temperatures the specimen relaxed and began to yield. Fractures in the restrained specimens developed parallel to the main axis of stress. Subsequent to fracture formation, the richest oil shale became so pliable during heating to 725° F. that it partially deformed under its own weight. A considerable portion of this oil shale, when heated to 825° F., exhibited free flow.

Extensive structural breakdown in the stress-free oil shales because of heat points to a mechanically weak mineral matrix. This indicates that the organic matter is the continuous phase and the predominant contributor to the oil shales' mechanical strength. The structural behavior of rich oil shales suggests that their properties may largely be those of the organic matter. Respective volume per cents of the organic matter in the three oil shales, calculated according to Smith (*9*), were 39.6, 48.5, and 60.0%, which also indicates that the properties of these oil shales may be primarily those of the organic matter.

Response of Cores to Heat and Uniaxial Stress. Yield temperatures of the specimens heated under compressive stress of 80, 200, or 325 p.s.i. are presented in Table I. The yield temperature denotes the temperature at which the specimen failed to support the load and underwent compressive strain. Immediately after the yield temperature was reached, compressive strain was slow; however, it accelerated with time and became very rapid as the specimen approached failure by fragmentation. At the yield temperature and above, the specimens were pliable and could be easily sliced with a knife either parallel or perpendicular to the bedding plane. The specimens exhibited considerable elastic rebound over a wide temperature range. This was determined by repeatedly reducing and then reapplying the uniaxial stress. The specimens appeared to exhibit simultaneously, in varying degrees, elastic and viscoelastic properties along with plastic deformation. Cores, which were cooled to ambient temperature after they reached the yield temperature, regained considerable mechanical strength with corresponding loss of pliableness and resilience.

Table I. Yield Temperature of Stressed Cores, ° F.

Stress, P.S.I.	Core Grade, Gal./Ton 34.5	45.5	63.5
80	737	727	721
200	709	704	686
325	690	685	658

After the specimens reached 725° F., the minimum and maximum time intervals to structural failure by fragmentation were 12 minutes for the richest oil shale stressed to 325 p.s.i. and 2 hours and 15 minutes for the 34.5-gallon-per-ton oil shale stressed to 80 p.s.i. Loss of organic matter at the yield temperature varied from 3.5 to 7.5 weight % of the initial organic matter. These losses were probably bitumen, benzene-soluble material, rather than thermally degraded kerogen. Respective losses of organic matter at the point of structural failure were 11.6 and 22.3 weight %.

Several factors strongly indicate that the organic matter is both the continuous phase and the predominant contributor to the oil shales' mechanical properties and structural responses to heat and stress: the great difference in compressive strengths between the oil shales at ambient temperature and at 725° F. (about 10,000 and 325 p.s.i.); the ease with which these specimens could be sliced at 725° F., either perpendicular or parallel to the bedding plane; the fairly high degree of elastic rebound over a wide temperature range; and the extensive structural breakdown due to heat in a stress-free environment.

Figure 4,*B* through *D*, illustrates the specimens just prior to structural failure by fragmentation. As the specimens began to yield, minute fractures developed parallel to the main axis of stress. These small fractures, in turn, propagated and enlarged with continued loss of cohesion. Uniaxial stress was reduced just prior to failure to photograph the specimens; otherwise they would have collapsed. After cooling, the fragments withstood considerable stress, which indicated that the kerogen had lost much of its pliableness.

Response of Fragments to Heat and Stress. Structural response of a column of fragments to heat and stress and the effect of deformation on the induced porosity and permeability are presented in graphic form in Figures 5 through 8. Each figure consists of two related curves: The first presents compressive strain and the second presents permeability (cubic inches per square inch of surface per minute) both as a function of time.

The columns of fragments began to yield below 150° F. and continued to compress to about 400° F. At this point the fragments ceased to yield under the applied stress for 2 to 4 hours, as noted by the plateaus in each of the compressive strain-time curves. Below 400° F., compressive strain varied from less than 1% to 10% of the initial column height. These strains were attributed to reorientation of the particles and not to any significant loss in the support capability of the column. The preliminary compressive strains were probably due to heated bitumen and kerogen serving as a lubricant, which decreased the coefficient of friction between the fragments' points of contact. Swelling may also have influenced reorientation. The extent of the preliminary strain was a function of both the richness of the specimen and the applied force.

As heating continued, each specimen reached its yield temperature, at which time it could no longer support the applied stress. The well-defined transition points reflect a pronounced change in the oil shales' mechanical strength. The small loss of organic matter at the transition point—only 3.6 weight %, for example, for the 45.5-gallon-per-ton oil shale stressed at 325 p.s.i.—suggests that the abrupt

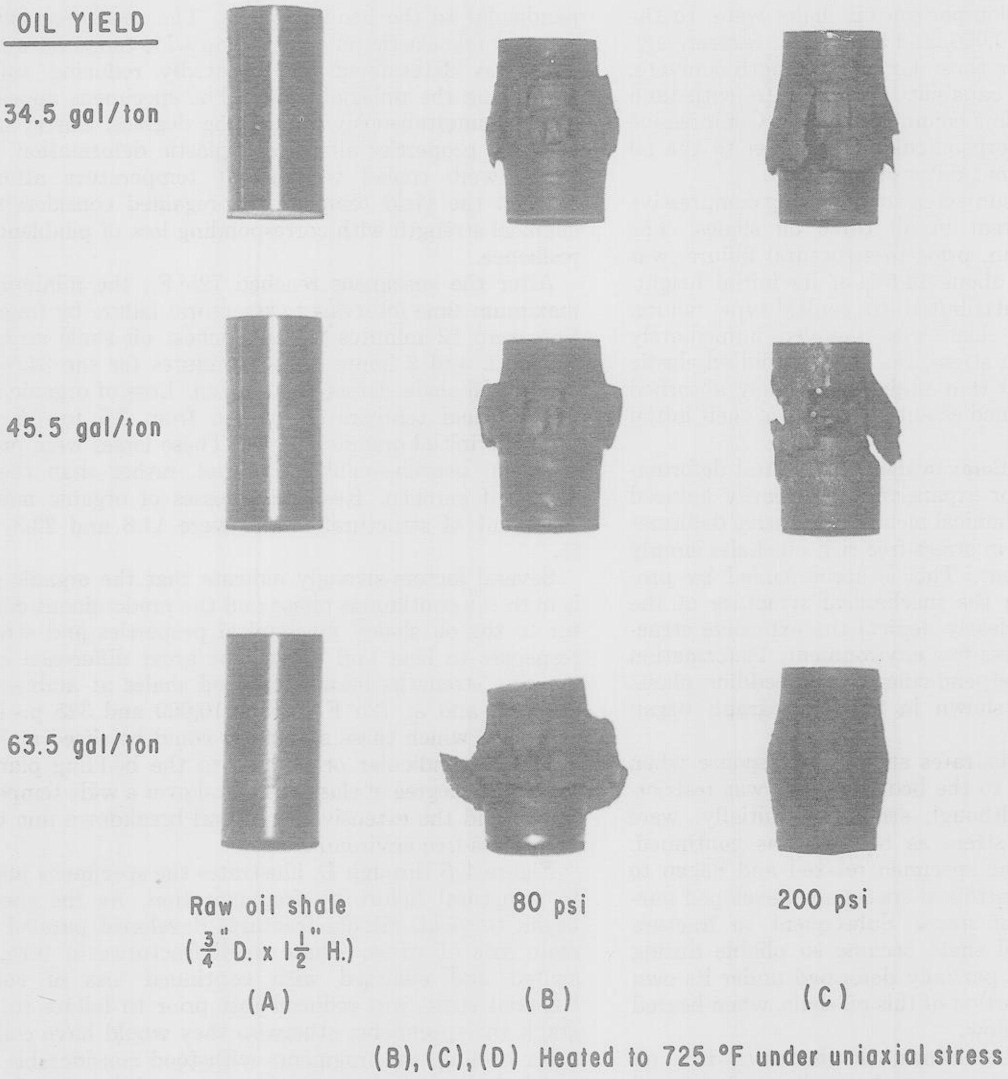

Figure 4. Structural response of oil shale cores to heat and compressive stress

change is primarily a physical instead of a chemical change. As did the stressed cores, the columns of fragments also exhibited elastic rebound and viscoelastic properties. As compressive strain increased with time, the columns' pore geometry, porosity, and permeability were concurrently undergoing continual change.

Under all three compressive stresses, the richest oil shale softened to the extent that both the mineral constituents and the organic matter extruded between the cell wall and the piston (clearance about 1 mm.). For this reason the tests for the 63.5-gallon-per-ton oil shale were terminated after about 8 hours. The equivalent spherical diameters of the mineral particles in very rich oil shale are predominantly less than 50 microns (*13, 14*) and, therefore, they could readily pass between a 1-mm. clearance. Time required for complete collapse after reaching 725° F. was stress-dependent: 2 hours and 50 minutes and 1 hour and 40 minutes for the 80- and 325-p.s.i. stresses, respectively. Tests for the other two oil shales were continued for several hours after maximum compressive strain was reached.

Effects of Heat, Stress, and Time on Permeability. Permeability in the permeability-time curves shown in Figures 5 through 7 is expressed as the amount of nitrogen (s.t.p.) through the column of fragments in cubic inches per square inch of surface per minute at a gas pressure differential across the column of fragments of 3 p.s.i. Permeabilities greater than 20 cu. inch per sq. inch of surface per minute were not measured. In most tests, permeability was not seriously impaired until the column of fragments had compressed at least 35% of its initial height.

FRAGMENTS FROM 63.4-GALLON-PER-TON OIL SHALE. The support capability of the column of fragments at 725° F. deteriorated to the extent that permeability was reduced

Table II. Experimental Data Relating to 63.5-Gallon-per-Ton Oil Shale Heated to 725° F.

Stress, P.S.I.	Compression at Zero Permeability, %[a]	Time Elapsed to Zero Permeability, Min.[b]	Time Elapsed to Structural Collapse, Min.[b]	Maximum Compression, %[a]	Organic Matter, Wt. % Loss
80	37.5	85	170	44.5	34.4
200	36.6	55	140	44.1	33.0
325	33.8	22	100	41.5	28.9

[a] Values represent per cent of initial column height. [b] Time measurements began at time specimens reached 725° F.

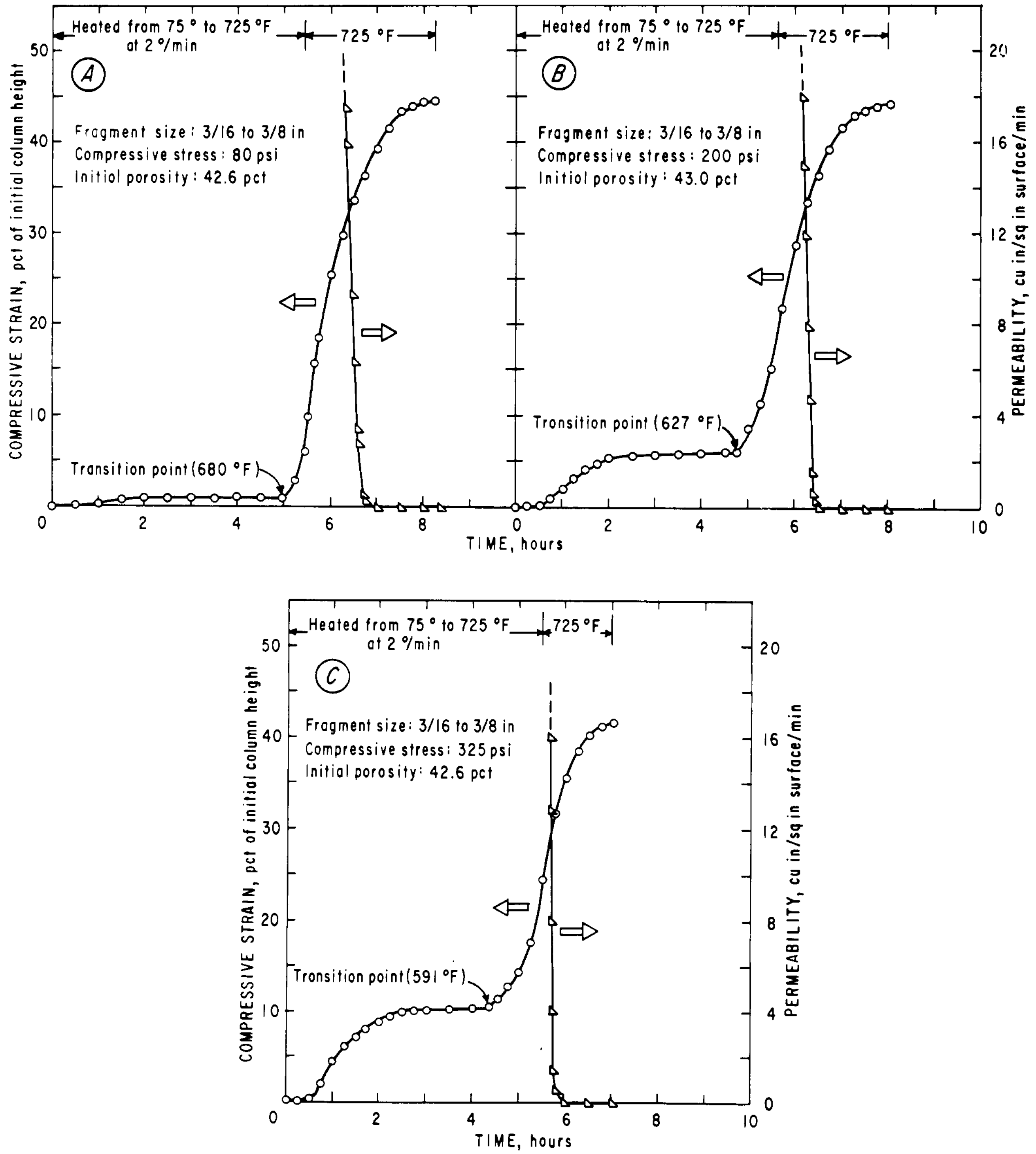

Figure 5. Compressive strain-time and permeability-time curves for 63.5-gallon-per-ton oil shale fragments

to zero under all three stress levels, as shown in Figure 5,*A* through *C*. Permeability reduced to zero within 22 minutes after the specimen stressed to 325 p.s.i. reached 725° F. Once permeability was lost, it was never restored under a pressure differential across the column of fragments at either 3 or 14.7 p.s.i. Table II presents data relating to compressive strain, permeability, time required for total collapse after reaching 725° F., and loss of organic matter at the time of collapse.

FRAGMENTS FROM 45.5-GALLON-PER-TON OIL SHALE. Permeability in the column of fragments stressed to 80 p.s.i. (Figure 6,*A*) remained above zero throughout the test. Minimum permeability of nitrogen during the 15-hour test was 0.43 cu. inch per sq. inch of surface per minute. Nitrogen was replaced with air at the end of this period. Sufficient permeability existed to oxidize the organic matter completely.

Compressive strain of the specimens stressed to 200 p.s.i. reduced permeability to zero. A typical permeability-time curve is shown in Figure 6,*B*. In this test permeability was reduced to zero after the specimen had compressed 35.2% of the initial column height. This particular test was continued beyond 15 hours to observe the oil shale's behavior. After 16 hours at a pressure differential across

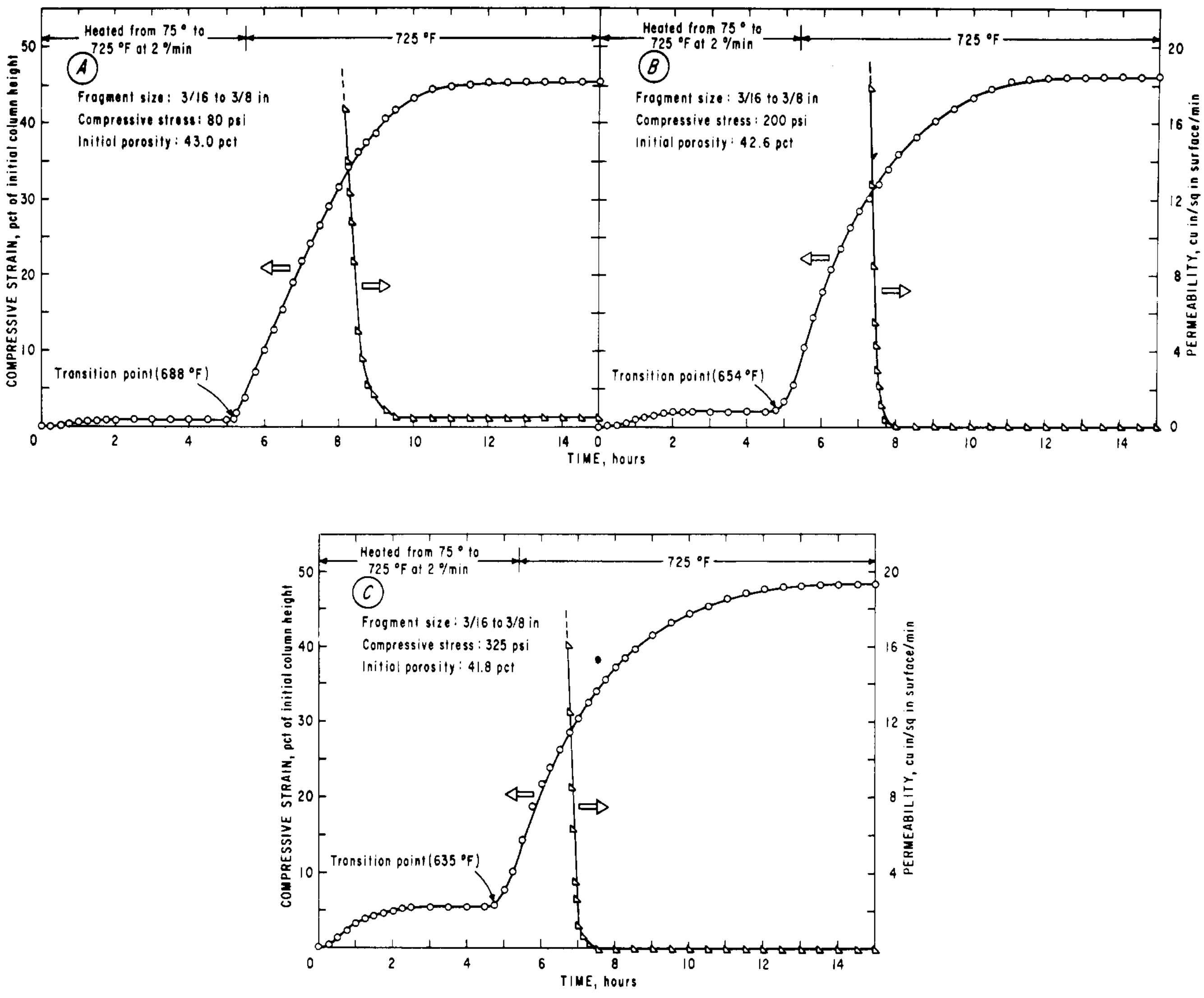

Figure 6. Compressive strain time and permeability-time curves for 45.5-gallon-per-ton oil shale fragments

the column of 3 p.s.i., permeability remained at zero. At this time the pressure differential was increased to 14.7 p.s.i. for an additional 12 hours. Some permeability was restored, and at the end of the test it amounted to 0.90 cu. inch per sq. inch of surface per minute.

Permeability was also reduced to zero in the fragments stressed to 325 p.s.i. A permeability-time curve for one of the tests is shown in Figure 6,*C*. Permeability was reduced to zero after the column of fragments compressed 35.5% of its initial height. During the 15-hour test, permeability remained at zero for 7½ hours. It also measured zero at a pressure differential across the column of 14.7 p.s.i. In one test a pressure differential of 14.7 instead of 3 p.s.i. was applied across the column of fragments immediately after permeability had reduced to zero. After 4 hours, permeability was partially restored, and at the end of 14 hours, nitrogen flow was 1 cu. inch per sq. inch of surface per minute. Nitrogen was replaced with air, and within several hours the organic matter was completely oxidized.

FRAGMENTS FROM 34.5-GALLON-PER-TON OIL SHALE. Permeability remained above zero in the specimens stressed to 80 and to 200 p.s.i. (Figure 7,*A* and *B*). At a pressure differential across the specimens of 3 p.s.i., the minimum flows of nitrogen during the 15-hour tests at these two stresses were 1.70 and 0.06 cu. inch per sq. inch of surface per minute after the columns of fragments had compressed 37.8 and 42.0%, respectively. After the 15-hour test, nitrogen was replaced with air. Sufficient permeability existed to oxidize the organic matter completely.

In the column of fragments stressed to 325 p.s.i. (Figure 7,*C*), permeability was reduced to zero after the column of fragments had compressed 40.5%. The test was continued beyond the 15 hours shown in Figure 7,*C*. Sixteen hours after zero permeability was reached, nitrogen flow was 0.12 cu. inch per sq. inch of surface per minute at a pressure differential across the column of 3 p.s.i. Nitrogen was replaced with air and the test continued for an additional 12 hours. At the end of the test period the carbonaceous matter was essentially intact, with no significant oxidation having taken place.

FRAGMENTS FROM 45.5- AND 34.5-GALLON-PER-TON OIL SHALES HEATED TO 825° F. The rate of compressive strain was greatly increased in the two oil shales heated to 825° F. as compared with those heated to 725° F. This is clearly shown in Figure 8,*A* through *C*. Maximum compressive strain occurred in about 1 hour after the specimens reached their transition point or yield temperature. Those heated

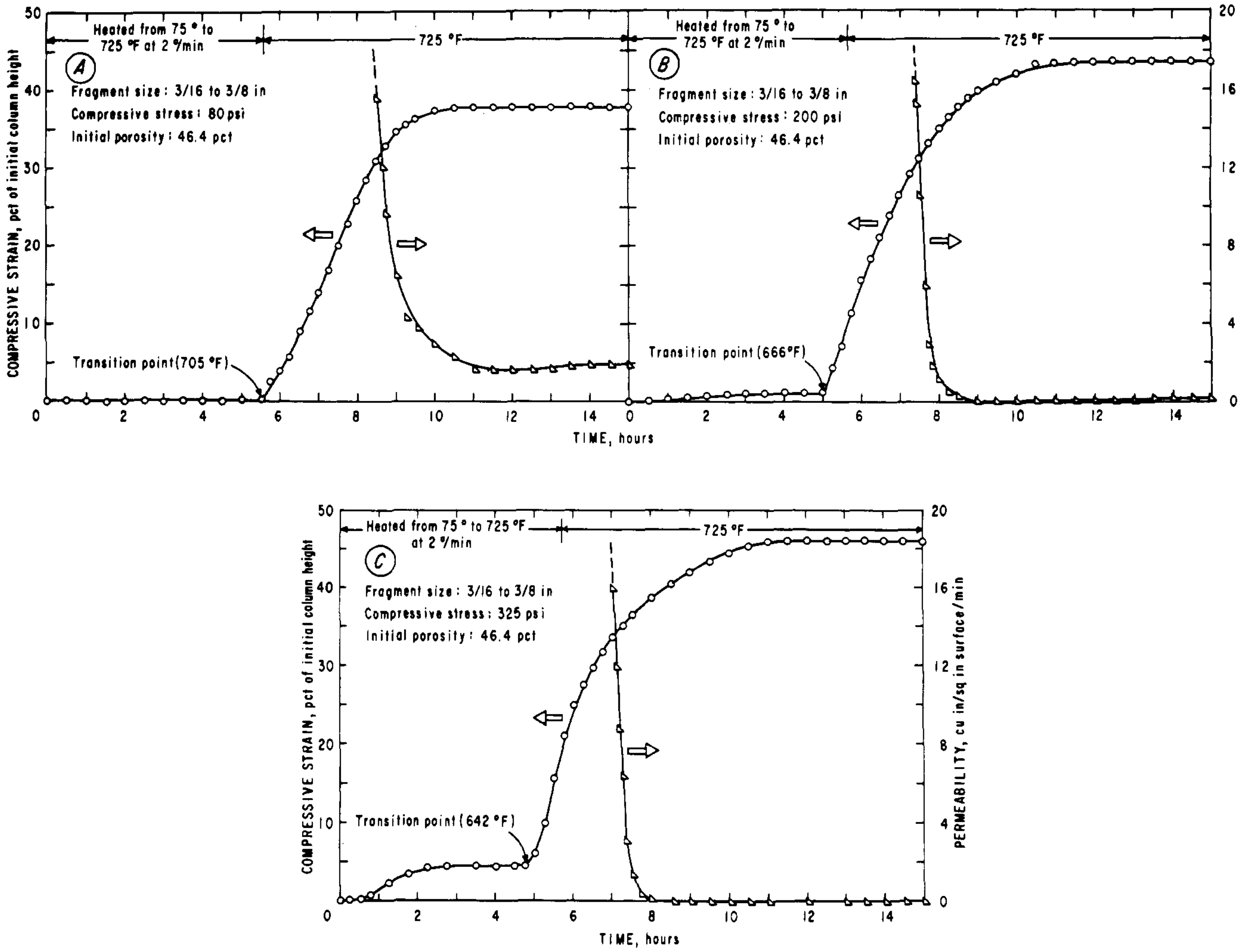

Figure 7. Compressive strain-time and permeability-time curves for 34.5-gallon-per-ton oil shale fragments

to 725° F. required several hours for maximum compressive strain to occur. Permeability was partially restored in each of the columns of fragments represented by Figure 8,*A* through *C*. In Figure 8,*A* and *B*, where the pressure differential across the column was 3 p.s.i., the respective permeabilities at the end of 12 hours were 0.52 and 0.93 cu. inch per sq. inch of surface per minute, and in Figure 8,*C*, where the pressure differential was 14.7 p.s.i., permeability had recovered to 5.74 cu. inch per sq. inch of surface per minute at the end of the test.

SUMMARY

Results from this investigation provide a better understanding of the structural response and/or deformation of small specimens of rich oil shales as they are heated to subretorting temperatures both in a stress-free environment and under moderate stresses.

Stress-strain-time-temperature relationships of the rich oil shales yielded data as to their yield temperature, yield stress, rate of compressive strain, structural deformation, loss of mechanical strength, and the effect of these properties on induced permeability.

Under the applied stresses, 80, 200, and 325 p.s.i., each oil shale exhibited a distinct yield temperature at each stress level. These temperatures were well below those required for rapid pyrolysis of the organic matter to produce oil. At their respective yield temperatures the oil shales became pliable and no longer supported the applied load. On cooling, however, they regained much of their initial mechanical strength. Loss of organic matter at the yield temperatures was low, 3.5 to 7.5 weight %. The loss was probably bitumen (benzene-soluble material) instead of thermally degraded kerogen. The small loss of organic matter together with the resumption, on cooling, of much of the oil shale's mechanical strength indicated that at the transition point kerogen primarily underwent physical instead of chemical changes.

Experimental data from this study indicate that kerogen is the continuous phase in rich oil shales and is the predominant contributor to their properties and to their response to heat and stress.

Small columns of fragments from the three rich oil shales investigated were also not self-supporting systems when heated to 725° and 825° F. under compressive stress of 80, 200, or 325 p.s.i. In most instances the induced permeability in the column of fragments was reduced to zero. In some tests permeability was partially restored and all of the organic matter could be oxidized.

This investigation shows that structural deformation in rich oil shales can be expected to occur ahead of the retorting zone. Structural response of large masses of rubble of unknown size and size distribution cannot be accurately predicted from results on small specimens. The preliminary

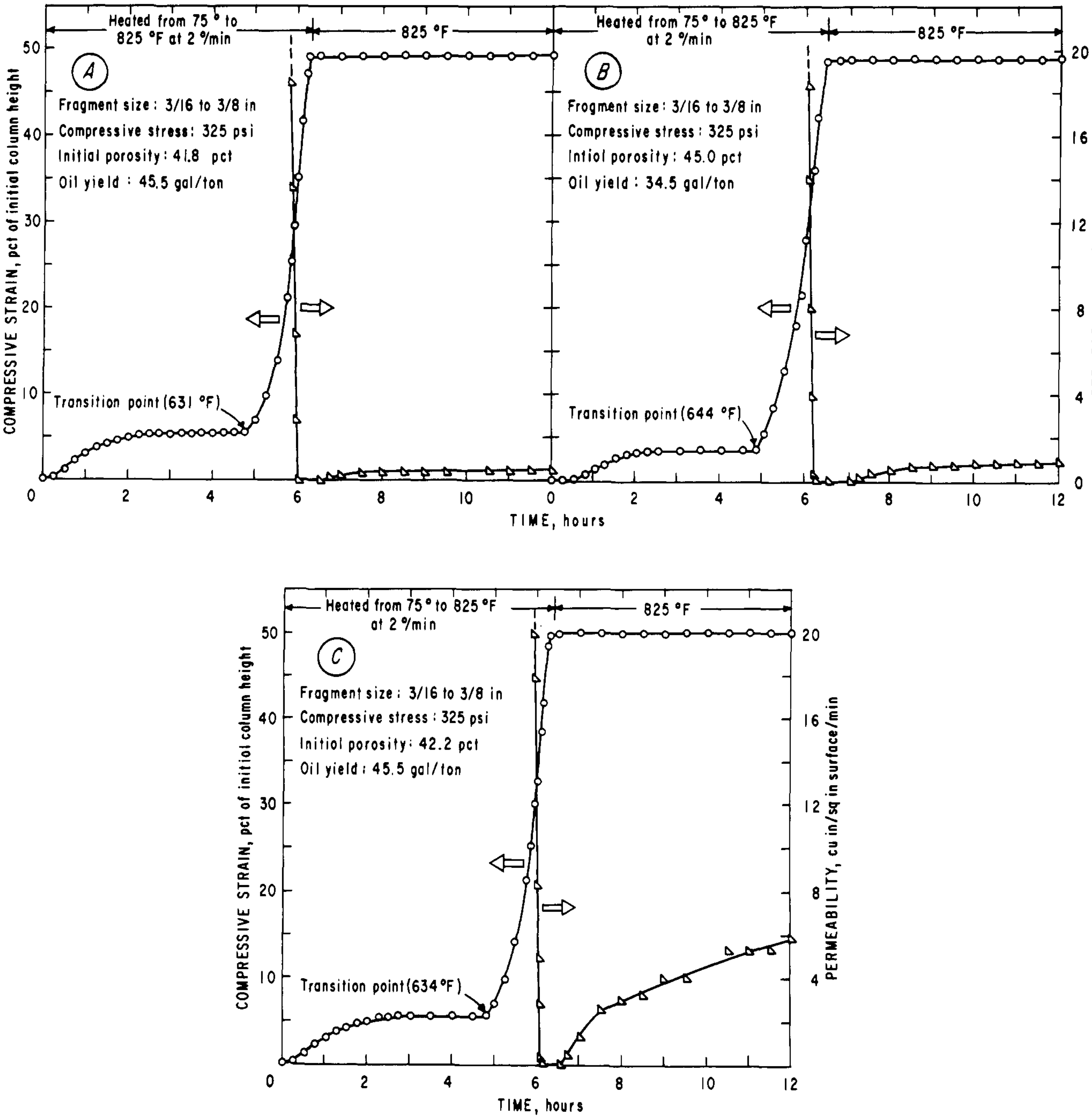

Figure 8. Compressive strain-time and permeability-time curves for 45.5- and 34.5-gallon-per-ton oil shale fragments

research, however, indicates that structural response of rich oil shales as they are heated under compressive stress may not be completely disregarded as a factor influencing underground retorting and it points to the need for additional engineering data to supplement the laboratory findings.

LITERATURE CITED

(1) Hull, W.Q., Guthrie, B., Sipprelle, E.M., *Ind. Eng. Chem.* **43**, 2 (1951).
(2) Institute of Petroleum, London, "Oil Shale and Cannel Coal," p. 345, 1951.
(3) Lekas, M.A., Carpenter, H.C., *Colo. School Mines Quart.* **3**, 7 (1965).
(4) Lombard, D.B., *J. Petrol. Technol.* **1965**, 877.
(5) Lombard, D.B., Carpenter, H.C., *Ibid.*, **1967**, 727.
(6) McKee, R.H., "Shale Oil," Chemical Catalog Co., New York, 1925.
(7) Matzick, A., *et al.*, Bur. Mines Bull. **635** (1966).
(8) Murphy, W.I.R., Proceedings of 2nd Plowshare Symposium, University of California, Lawrence Radiation Laboratories, **UCRL-5678**, 80 (May 1959).
(9) Smith, John Ward, Bur. Mines Rept. Invest. **7248** (1969).
(10) Smith, J.W., Thomas, H.E., Trudell, L.G., Society of Professional Well Log Analysts, 9th Annual Logging Symposium, June 23, 1968.
(11) Sohns, H.W., 13th Annual Field Conference, Intermountain Association of Petroleum Geologists, p. 223, 1964.
(12) Tisot, P.R., J. Chem. Eng. Data **12**, 405 (1967).
(13) Tisot, P.R., Murphy, W.I.R., *Chem. Eng. Progr. Symp. Ser.* **61**, 25 (1965).
(14) Tisot, P.R., Murphy, W.I.R., J. Chem. Eng. Data **5**, 558 (1960).

RECEIVED for review October 13, 1969. Accepted March 2, 1970. Division of Petroleum Chemistry, 158th Meeting, ACS, New York, N.Y., September 1969. Work done under a cooperative agreement between the Bureau of Mines, U.S. Department of the Interior, and the University of Wyoming.