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ANALYSIS OF HIGH PRESSURE OIL SHALE HUGONIOTS

MASTER

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ANALYSIS OF HIGH PRESSURE

OIL SHALE HUGONIOTS

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ABSTRACT

A collection of low and high stress level Hugoniot data for a low kerogen content, porous Anvil Points oil shale are analyzed with the P- α model to give a Hugoniot for kerogen free shale. The P- α model suggests an "elastic" yield stress, P_e , of 0.5 GPa and a solid density complete compaction pressure, P_s , of 10.0 GPa. Knowledge of this behavior permits the calculations of the in situ, "effective", Hugoniot of kerogen from extensive Hugoniot data for various mixtures of shale and kerogen. The Hugoniot of kerogen free shale is $U_s = 4.30 + 1.27 u_p$ (km/s); $\rho_o = 2.65 \text{ Mg/m}^3$ and of kerogen is $U_s = 3.09 + 1.16 u_p$ (km/s); $\rho_o = 1.05 \text{ Mg/m}^3$, where U_s is the shock velocity, u_p the particle velocity and ρ_o the initial density.

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INTRODUCTION

As a portion of the modeling program for predicting the dynamic response of oil shale, we want to look at some of the very simple Hugoniot models. While the long range goals for adequate modeling of wave propagation and fragmentation will undoubtedly require more sophisticated constitutive descriptions, at this early stage of the program much can be gleaned from a thorough analysis of simple data. Fortunately, a large body of experimental Hugoniot data on Green River oil shale from the Anvil Points mine is available for analysis. The materials examined have a range of kerogen contents.¹ Although the stress levels of the study were much larger than the range of stresses expected in the in situ bed preparation field experiments, appropriate extrapolations to lower stress levels are possible. Also, a few experimental data in the appropriate stress range on a similar Anvil Points material can be obtained from the work of Schuler et al.²

The thrust of our analysis of these data differs from previous analyses in the details of the models used and also in the approach to the use of the experimental data for deducing unknown properties of the components of the oil shale material. Initially, the Hugoniot data on a very lean, but porous, oil shale are used to obtain a Hugoniot of the kerogen free, nonporous shale material. Then the Hugoniots of this kerogen free shale and of other shales with various kerogen contents are used to deduce an "effective" Hugoniot of the kerogen. The kerogen Hugoniot is "effective" because it represents the modified response of the kerogen in place as contrasted to the response of bulk kerogen. While this precludes our making estimates of the influence of material strength on the dynamic response, pragmatically, it does lead to a better description of the kerogen response for calculational purposes.

ANALYSIS OF SOLID DENSITY SHALE HUGONIOTS

In terms of the dynamic response, the oil shale can, to a first approximation, be considered as a porous shale matrix material in which the pores may or may not be filled with kerogen. It appears at the higher densities, where the kerogen contents are very small, that much of the pore volume tends to be unfilled by kerogen. Our knowledge of porous material response can be used to determine from these porous materials the most probable Hugoniot of kerogen free, solid density shale.*

Hugoniot Response Data of Shale:

The extensive data of Carter¹ for oil shale contains a data set on a nearly kerogen free (less than 3 gal/ton) shale with 11.8% porosity by volume. Porosity was determined by aspiration techniques using water. Unfortunately, only a few data points are in the appropriate stress region below the dolomite phase transformation.³ While this introduces some uncertainty in the analysis, a suitable Hugoniot can be obtained. Carter has already analyzed these data on the basis of a simple collapse model, and we will not repeat that process. The solid, kerogen free, shale Hugoniot obtained by Carter is given by**

* The mineral components and their volume fractions are additional variables which would modify the Hugoniot response, we assume here that these remain effectively the same for all oil shale.

** Because of the limited data involved, we found it possible to deduce, with his same simple collapse model, somewhat different results than Carter's; however, the differences were not really significant.

$$U_s = 4.30 + 1.27 u_p ; \rho_o = 2.65^*$$

where U_s is the shock velocity and u_p the particle velocity in km/s and ρ_0 the initial density in Mg/m³.

In the ensuing calculations, the equation of state of the solid is taken to be the Mie-Gruneisen equation. The value of the Gruneisen parameter, γ , is taken as 1.5. An appropriate value of γ is necessary for the calculations; however, within reasonable limits the calculations are somewhat insensitive to the exact value chosen. As is often done, the product, $\rho\gamma$, is taken as a constant.

We make use of this solid density Hugoniot as an input to the P- α model⁵ of porous material response to calculate the observed behavior. This perhaps seems a circular argument; however, it will become clear that additional knowledge of the material response is deduced in the process. The P- α model relates pressure states of the solid to pressure states of the distended material at the same internal energy through a distention ratio α . Here α is defined as the ratio of porous to solid densities and is assumed to be a quadratic in pressure between P_e , the elastic pressure, and P_s , the pressure of complete pore closure. The appropriate integrations to accomplish this process are given in the original reference.

* Carter used a density of 2.65 Mg/m^3 ; presumably calculated from the volume fractions of mineral constituents and their individual densities. Composition of this material was found by x-ray analysis. If we calculate theoretical densities for all the "whole shale" samples analyzed by Olinger,⁴ the average density of the 13 samples is $2.654 + .073 \text{ Mg/m}^3$,
- .041

even though concentrations of individual constituents vary considerably among the samples.

Calculations using the P- α model can give results for a simple collapse model by letting $P_s = P_e = 0$; this duplicates Carter's analysis.¹ For the Carter material with 11.8% porosity by volume, the initial distention ratio, α_0 , is 1.133. The results of this analysis, together with the experimental data, are shown in Fig. 1. In the figure, the dashed line labeled $P_s = P_e = 0$ merges with the other calculations above the U_s , u_p values corresponding to P_s in the other calculations. As is apparent, this calculation is in agreement with only the last high pressure datum. The significant deviation at lower particle velocities is expected and results from a resistance of the material to collapse. Although we have insufficient data to describe this resistance, educated guesses of the appropriate values for P_s and P_e can be made. In this case, a value of 10 GPa for P_s , which is reasonable for relatively strong materials such as shale, gives a calculated curve which nicely agrees with all of the Carter data, see the solid curve in Fig. 1 labeled $P_s = 10$ GPa. (Note that a small portion of the curve generated by this calculation is dashed at very low particle velocities).

The rather sparse high pressure data are augmented by a collection of direct impact data at about 1.4 GPa. Further analysis of the raw quartz gauge records* used by Schuler et al.² permits calculation of Hugoniot points. These experiments were on the same** low kerogen content, porous oil shale as Carter used; however, the porosity determined by Schuler et al. using a helium picnometer on fine powder was 9% by volume.

* Lysne performed these experiments and made the records available to the author for the analysis.

** The work at Los Alamos Scientific Laboratory was apparently done on material from the same block of shale as the Sandia work.

The records were from an in-projectile quartz gauge which recorded the impact stress as the gauge impacted the oil shale target. From the impact stress, the projectile velocity and the constitutive equation for single crystal quartz, one can calculate the particle velocity, u_p , and shock velocity, U_s , for the oil shale. The impact gauge records were not straightforward, in that they contained evidence of a high initial stress, followed by stress relaxation to a plateau, and then an increase of stress with considerable ringing or noise. We believe this to be a result of the porosity.⁶ As a consequence, the long time or Hugoniot response of the material was taken as the plateau stress averaged through the ringing. Analysis of these tests using these stresses for the calculations gives the results in Table I. Average values for the three orientations are plotted in Fig. 1. A P- α model calculation at 9% porosity, $\alpha = 1.098$, and $P_s = 10$ GPa is shown in the figure for comparison to the data. Agreement is satisfactory.

A great deal of information can be obtained from existing data on the small signal response of this oil shale. Schuler *et al.*² give first arrival velocities of transmitted stress waves. The conditions approximate $u_p = 0$ and the results are repeated in Table I. Average values for the three orientations are plotted in Fig. 1. The open squares represent the 0° orientation, in which the bedding planes are normal to the wave propagation direction. The partially filled squares are for 45°, and the filled squares for 90°.

There appears to be some influence of specimen orientation on the first arrival and Hugoniot velocities. While the 8% differences in velocities between extremes are probably within the experimental uncertainty*,

*The quality control on the selection of specimens and specimen uniformity for this early work was less informed than current practice.

TABLE I - HUGONIOT DATA - ANVIL POINTS
(Low Kerogen Content - Porous Material)

Designation	Density (Mg/m ³) ρ_0	Ultrasonic Velocities (km/s)			Shock Wave*			
		C_L	C_O	C_S	Initial Wave Velocity (km/s) U_i	Particle Velocity (km/s) u_p	Hugoniot	
							Shock Velocity (km/s) U_s	Average Pressure (GPa) P
Schuler et al. ² (9% porosity)								
0° Orientation								
Shot 2	2.38				4.36	.1745	3.347	1.39
5	2.31				4.15	.1705	3.478	1.37
7	2.34				-	.1724	3.298	1.33
9	2.38				4.39	.1784	3.133	1.33
Ave					4.30	.1739	3.314	1.35
45° Orientation								
Shot 1	2.35				4.36	.1761	3.117	1.29
4	2.39				4.30	.1754	3.077	1.29
6	2.37				4.40	.1735	3.186	1.31
10	2.37				4.54	.1698	3.430	1.38
Ave					4.40	.1737	3.203	1.32
90° Orientation								
Shot 3	2.35				4.68	.1700	3.629	1.45
8	2.35				4.69	.1668	3.622	1.42
11	2.37				4.42	.1662	3.631	1.43
12	2.37				4.66	.1712	3.381	1.36
Ave					4.61	.1686	3.566	1.42
Olinger ³ (11.8% porosity)								
0° Orientation								
Dry	2.22	4.03	3.20**	2.47				V_1 & V_4
Saturated	2.33	3.66	3.18**	1.99				V_1 & V_4
45° Orientation								
Dry	2.22	4.28						V_5
Saturated	2.33	3.90						V_5
90° Orientation								
Dry	2.22	4.44						V_1
Saturated	2.33	4.01						V_1
Schuler et al. ² (9% porosity)								
0° Orientation	2.40	4.22						V_1
45° Orientation	2.39	4.50						V_5
90° Orientation	2.30	4.88						V_3

* The analysis technique is that given by Munson⁵ and was used on the raw data records.

**

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* The analysis technique is that given by Munson⁵ and was used on the raw data records.

** Calculated from the transversely isotropic moduli using formulas given by Schock et al.⁶

the trend is in accord with anisotropy effects in composites. There is a reversal of the 0° and 45° orientation velocity positions between the Hugoniot and first arrival data. It is doubtful that any significance should be attached to this reversal. A comment on the magnitude of the velocity variation with orientation is necessary. The 8% variation in velocity could be an indication of anisotropy, which one would anticipate for oil shale. The magnitude of the effect amounts to approximately a 17% change in modulus.

Interpretation of the large first arrival velocities requires some care. Although this could be interpreted as an elastic precursor, there is little evidence in the transmitted wave profiles of such an effect. Such behavior can be treated in the P- α model by an additional feature of the crush-up curve; specifically, the incorporation of a greater resistance to collapse of the voids during early stages of compaction. When this is done, an additional variable, P_e , analogous to the onset of crushing is introduced. For pressures less than this value, the material has a response called "elastic" in the sense deformation is assumed recoverable. As an illustration we show in Fig. 1 a calculation for an assumed value of $P_e = 0.5$ GPa and C_e , sound speed, of 4.5 km/s (this is the sound speed associated with the virgin porous material and in this case has a value greater than the bulk velocity of the solid Hugoniot). The influence of P_e on the calculation is apparent at the low particle velocities where it causes the shock velocity to increase abruptly from the values calculated without this parameter. In the figure the solid curves, as is now evident, are the calculations with both P_e and P_s included. While a discrete value of P_e would produce a step function precursor in the stress wave not experimentally observed, schemes to introduce more complicated initial

crush-up behavior through functional forms of P_e were not really warranted. We can see features with a single value P_e which are basically correct. The calculation agrees well with the initial arrival velocities at zero particle velocity of the shock wave experiments of Schuler et al.²

Ultrasonic Data - Specific to the Analysis:

Ultrasonic velocity determinations, providing the Hugoniot analysis is appropriate, should verify important intercept values for the Hugoniot of both the porous material response and the deduced solid material response. The most extensive ultrasonic work is that of Olinger⁴ using not only a similar low kerogen, high porosity oil shale similar to that tested by Schuler et al.² and Carter¹ but also numerous other materials with various kerogen contents.

On the low kerogen, porous Anvil Points material, Olinger⁴ measures ultrasonic velocities for dry and water-saturated specimens. These correspond to a transversely isotropic material. We give in Table I, the quasi-longitudinal equivalent of 0°, 45° and 90° orientations which are designated as V_3 , V_5 and V_1 , respectively. The various dry condition quasi-longitudinal velocities, while somewhat less (6.3, 2.7 and 3.7%, respectively) than the first arrival velocities of Schuler et al.², are still well within acceptable limits of agreement. Also from these data, a calculated bulk modulus gives 3.20 (dry) and 3.18 (saturated) km/s for bulk wave speeds.* The intercept of the extrapolated P - α calculated Hugoniot for $P_s = 10.0$ GPa is in very good agreement with this ultrasonic bulk velocity, an agreement which is necessary for consistency of the material response.

* In making the calculations of bulk moduli; hence the bulk velocities, C_o , we have used the transversely isotropic relationships given by Schock et al.⁷ As a consequence, these values may differ somewhat from the originally reported values.

The ultrasonic data of Olinger⁴ are very extensive, with the five ultrasonic velocities being determined as a function of oil shale bulk density over the range of 1.4-2.37 Mg/m³. His analysis suggests each velocity can be represented as a function of density by a bisegmented, piecewise linear relationship. The two segments are connected at a density of 2.0 Mg/m³, the theoretical density of in-contact shale spheres. The linear fits are reproduced in Fig. 2.

Since there is a monotonic relationship between the density and the kerogen content, these wave speeds can be taken as a representation of kerogen content directly.⁸ Olinger⁴ used extrapolation of these data to a "nonporous" density of 2.512 Mg/m³ to calculate bulk moduli for the low kerogen content material. While the concept of this extrapolation is proper, we believe that the extrapolation should have been to the theoretical, kerogen-free, shale density of 2.65 Mg/m³. Our reasoning is that the correlation is in kerogen content and density, not porosity and density. In fact, the influence of porosity would be to cause negative deviations from the linear relationship between velocity and density (kerogen content). This effect would first become increasingly strong at the higher densities as the probability of significant porosity becomes greater and then would decrease again as the density approached the theoretical solid shale density, i.e. both the void free, sonic velocity vs. density and the porous, sonic velocity vs. density curves will extrapolate to the same point, i.e. the velocity for kerogen free shale of theoretical density. Thus, the ultrasonic velocities for solid shale are, by extrapolation, $V_1 = 6.22$, $V_3 = 5.44$, $V_4 = 3.22$, $V_5 = 5.74$ and $V_6 = 3.81$ km/s (see Fig. 2). Calculation of a bulk modulus permits a bulk sound speed of 4.30 km/s to be determined which agrees with the intercept

of the Hugoniot deduced by Carter. One should note that this value of bulk sound speed differs from that of the solid density shale Hugoniot deduced from the bulk moduli and densities by Olinger because the extrapolation was to a different density.

The bulk sound speed of 4.30 km/s is an additional verification of the proper selection of the kerogen-free, solid density shale response. Use of the P- α model and all available Hugoniot and ultrasonic data, form a consistent picture of the Hugoniot for the low kerogen porous shale. This consistent picture of the porous material response through the model and with support of extensive ultrasonic and density data yields a very reasonable description of the solid shale Hugoniot. The result of our more detailed reanalysis supports the Hugoniot used by Carter initially.

Ultrasonic Data - General:

Although the Olinger data are the most complete set available, several less extensive collections are known. It is of interest to compare these data because they represent not only similar Anvil Points material but also other Green River Shales, including some Rock Springs Site 9 materials.

As a preliminary to Sandia's participation in Rock Springs Site 9 activities, longitudinal (V_3) and shear (V_4) velocity data were obtained on available Anvil Points specimens and longitudinal (V_3) velocity data were obtained on core samples from Rock Springs Site 9 Gauge Hole #1 at the 43.3-43.9 meter depths. These unreported data⁹ were obtained by Schuler and Stevens.* Some of the Anvil Points material used here was the same as that used by Schmidt and Schuler for compressive failure surface tests.¹⁰ The remainder was that tested by Schuler et al.² These Anvil

* Schuler and Stevens made these data available for inclusion here.

Points data are shown in Fig. 2, and are in agreement with the data of Olinger.⁴

The data on the Rock Springs material does not correlate well with Olinger's data. As given in Fig. 3, Schuler and Stevens' data on the Rock Springs material show considerable scatter and a less distinct increase in velocity with increases in density. The observation is supported by the complete data set of Johnson¹¹ on the same core material. Also included in the graph are the data of Schock et al.⁷ on a Green River Shale from 704.1 m depth in well hole CCH-3, Rifle, Colorado. These data suggest significant variations in material moduli. Eventually, the cause for these variations must be found and correlated to the dynamic response.

HUGONIOT RESPONSE OF KEROGEN

Dremin and Karputkin¹² have shown that the Hugoniot of a constituent of a mechanical mixture can be obtained from the Hugoniots of the mixture and the other component. We make use of this analysis to deduce the "effective" Hugoniot of kerogen. On the basis of independent information, the Hugoniot of kerogen free shale has been obtained. Using this and Carter's¹ experimental data for mixtures of shale and kerogen, the analysis produced the results shown in Fig. 4. All data, save for the four very high particle velocity points, define a linear relationship of

$$U_s = 3.09 + 1.16 u_p \text{ (km/s)} ; \quad \rho_o = 1.05 \text{ Mg/m}^3 .$$

This Hugoniot for kerogen differs from the usual Hugoniot because it represents the response within the shale matrix rather than bulk material response. It may also include contributions to the mixture response from other sources, such as strength effects in the shale. In our analyses, we have taken the density of kerogen suggested by Carter.¹

Although no clear explanation of the discrepancy in the high particle velocity data is available, these data are in the stress range where a polymorphic phase change in dolomite may initiate.³ Other possibilities exist, and more information would be necessary to establish the cause.

Back calculation of the mixture response using the component Hugoniots was done to check our results. As shown in Fig. 5, the calculations represent, on the average, the experimental results, as they must.

We must emphasize again that the kerogen Hugoniot deduced here is not the bulk response of kerogen. It will deviate at low stress levels, especially.

SUMMARY

An analysis of Carter¹ and Schuler et al.² Hugoniot data in terms of the P- α model of porous material response suggests that one of the Anvil Points materials, extremely lean in kerogen, conforms well to this model. Values of $P_e = 0.5$ and $P_s = 10.0$ GPa reproduce observed data trends, and also show agreement with the bulk ultrasonic velocity. The analysis does not admit material anisotropy, although anisotropy is evident in the low stress level Hugoniot data.

The Hugoniot of kerogen free, solid shale initially proposed by Carter was found completely adequate for the analysis. It appears that bulk moduli based on simple calculations from extrapolations in density of the transversely isotropic response of the ultrasonic velocities are in agreement with the Hugoniot data. While it is difficult to know exactly the meaning of such bulk moduli calculations for anisotropic materials, the internal consistency is gratifying.

Comparison of two collections of Anvil Points ultrasonic velocity data indicated general agreement, even though the materials apparently were not from the same lot. However, there are certainly material variations which alter the ultrasonic response as demonstrated by the significant differences between Rock Springs and Anvil Points materials. Much work needs to be done to verify these differences, demonstrate their origin and formulate a predictive model describing the material effects.

Back calculation of the in place kerogen response from the solid shale and experimental mixture data was done. The "effective" Hugoniot deduced in this manner is

$$U_s = 3.09 + 1.16 u_p \text{ (km/s)} ; \rho_o = 1.05 \text{ Mg/m}^3$$

and is in general stiffer at low stress levels and less stiff at large stress levels than had been used in the work of Carter.

FIGURE 1. ANALYSIS OF POROUS LOW KERGEN CONTENT OIL SHALE USING P- α MODEL.

FIGURE 2. ULTRASONIC VELOCITY DATA - ANVIL POINTS OIL SHALES.

FIGURE 3. ULTRASONIC VELOCITY DATA - ROCK SPRINGS AND OTHER OIL SHALES.

FIGURE 4. HUGONIOT OF KEROGEN AS DEDUCED FROM MIXTURE THEORY.

FIGURE 5. MIXTURE CALCULATIONS FOR VARIOUS KEROGEN CONTENT OIL SHALES.

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