Viscosity Modeling and Prediction of Reservoir Fluids: From Natural Gas to Heavy Oils¹

S. E. Quinones-Cisneros, ˜ ²,³ **C. K. Zeberg-Mikkelsen, ´** ⁴ **A. Baylaucq,**⁴ **and C. Boned**⁴

> Viscosity and density are key properties for the evaluation, simulation, and development of petroleum reservoirs. In previous work, the friction theory (f -*theory*) models have already been shown capable of providing simple but accurate viscosity modeling results of petroleum reservoir fluids with molar masses up to around $200 g \cdot mol^{-1}$. As a base, the *f*-*theory* approach requires a compositional characterization procedure to be used in conjunction with a van der Waals type of equation of state (EOS). This is achieved using simple cubic EOS, which are widely used within the oil industry. In this work, the f -*theory* approach is further extended to the viscosity modeling of heavy reservoir fluids with viscosities up to thousands of mPa ·s. Essential to the extended approach presented here is the achievement of accurate *pvT* results for the EOS characterized fluid. In particular, it has been found that for accurate viscosity modeling of heavy oils, a compressibility correction in the way the EOS is coupled to the viscosity model is required. With the approach presented in this work, the potential of the f -*theory* for viscosity modeling of reservoir fluids is extended to practically all kind of reservoir fluids, from light ones to heavy ones. Additionally, the approach has been completed with an accurate density modeling scheme.

> **KEY WORDS:** characterization; density; equation of state; *f*-theory; modeling; petroleum; viscosity.

1353

¹Paper presented at the Fifteenth Symposium on Thermophysical Properties, June 22–27, 2003, Boulder, Colorado, U.S.A.

²Department of Chemical Engineering, Technical University of Denmark, Building 229, DK-2800 Lyngby, Denmark.

 3 To whom correspondence should be addressed. E-mail: seqc@mac.com

⁴Laboratoire des Fluides Complexes, Faculté des Sciences et Techniques, Université de Pau, Avenue de l'Université, BP 1155, 64013 Pau Cedex, France.

1. INTRODUCTION

In previous work, the friction theory (f -*theory*) [1] for viscosity modeling has been applied to the accurate viscosity prediction of light reservoir fluids (natural gas) [2] and accurate modeling and prediction of denser reservoir fluids, with molar masses up to around 200 g · mol⁻¹ [3]. However, viscosity modeling of heavy reservoir fluids, with molar masses up to more than 400 g · mol−¹ at reservoir conditions and with viscosities on the order of thousands of mPa ·s, remained untouched. Thus, while preserving most of the simplicity and accuracy of the original formulation, in this work, the f -*theory* approach is further extended to heavy reservoir fluids. Furthermore, the scheme illustrated here is still based on the same kind of simple cubic equations of state that are commonly used in the oil industry.

The *f*-*theory* for viscosity modeling consists of simple but accurate viscosity models that take advantage of the repulsive and attractive pressure terms in van der Waals type equations of state (EOS), such as the Soave–Redlich–Kwong (SRK) EOS [4] or the Peng–Robinson (PR) EOS [5] (PR). In subsequent work [6, 7], generalized f -*theory* one-parameter models have been developed to be used in conjunction with some of the most popular EOS. These models can accurately represent the viscosity of a large number of pure hydrocarbons and, using simple mixing rules and pure-component parameters, accurately predict mixture viscosities. The one-parameter f -*theory* models have been further extended to accurate viscosity prediction and modeling of characterized crude oils [3]. In an oil characterization procedure the light fraction, up to C_6 , is normally described in terms of 11 well-defined components: N_2 , CO_2 , H_2S , CH₄, C₂H₆, C₃H₈, *i*-C₄, *n*-C₄, *i*-C₅, *n*-C₅, and C₆. However, the heavy (C_{7+}) oil fraction is characterized in terms of a given number of pseudocomponents for which some characteristic scaling parameters are derived so that the phase behavior is correctly reproduced. In this work, a new characterization procedure [8], which is a generalization of previous work [9], is used.

2. OIL PVT CHARACTERIZATION

The core of the viscosity modeling work presented here is the oil characterization into a number of compounds and pseudo-compounds capable of, in conjunction with an EOS, providing adequate phase and viscosity behavior performance. The characterization procedure that is used in this work is based on the application of a probability distribution function for the representation of the heavy (C_{7+}) fraction. Such an approach was first proposed by Whitson [10] and it has been recently

reviewed by Whitson and Brulé in Chap. 5 of Ref. 11. The probability distribution function that is used in this work is the chi-square distribution function with p degrees of freedom $(CS(p))$, which is a particular case of the more general Gamma distribution function originally used by Whitson. However, among other minor points, the main difference between both methods is not the mathematical model used but rather the fact that the probability distribution function in the Whitson method is used to represent the molar fraction distribution while in the case presented in this work it is used for the mass fraction distribution. In terms of the mass fraction characterization approach used in this work, the procedure consists of characterizing the heavy oil fraction by distributing the C_{7+} mass fraction according to the $CS(p)$ function that best represents the mass distribution of the fluid. The general mathematical form of the $CS(p)$ distribution function is

$$
f_{\text{dis}} = \frac{2^{-p/2}}{\Gamma(p/2)} e^{-s/2} s^{(p/2)-1}
$$
 (1)

where s can be considered as a mass distribution variable. Here, s varies from zero to infinity but the light-component mass fraction, which corresponds to the integral from zero to an initial s_0 value, is excluded. Thus, it is required that

$$
M_6 = \int_{0}^{s_0} f_{\text{dis}} ds = \frac{1}{M W_{\text{T}}} \left(\sum_{i} x_i M W_i \right)
$$
 (2)

where M_6 represents the total fluid light mass fraction up to the C_6 fraction and s_0 is the value of s that satisfies Eq. (2). Clearly, the summation over i in Eq. (2) covers all light compounds in a given oil sample up to hexanes and MW_T is the total molar mass of the fluid. Then, the C_{7+} fraction is characterized as an m number of heavy fractions F_i of mass fraction fm_i according to

$$
fm_i = \int_{S_{i-1}}^{S_i} f_{dis} ds
$$
 (3)

Finally, the molar mass MW_i of the fraction F_i is given by

$$
MW_i = \overline{MW}\hat{s}_i
$$
 (4)

where \hat{s}_i represents the center of mass for the F_i fraction and \overline{MW} is a scaling value so that the total mass balance is satisfied. That is, \hat{s}_i is calculated with the equation,

$$
\hat{s}_i = \frac{1}{fm_i} \int_{s_{i-1}}^{s_i} s f_{dis} ds
$$
\n(5)

and, from Eq. (4) and a mass balance, it can be shown that \overline{MW} is given by

$$
\overline{MW} = \frac{MW^+}{(1 - M_6)} \sum_{i=1}^{m} \frac{fm_i}{\hat{s}_i}
$$
 (6)

where MW^+ is the C_{7+} fraction molar mass.

In the actual fluid characterization procedure, this approach is first used to find the optimal $CS(p)$ function by minimizing a mass weighted absolute deviation between the molar mass corresponding to each one of the experimentally reported mass fractions $(C_7, C_8, \ldots, C_{m+})$ and the molar masses calculated with Eq. (4). For this minimization it has been found that it is enough to only consider $CS(p)$ functions with p between 2 and 10 at steps of 0.5, that is, $p=2, 2.5, 3, \ldots$, 10. Then, using the optimal $CS(p)$ probability distribution function, the entire C_{7+} fraction is characterized in a sensible number of heavy fractions. In this work the C_{7+} fraction is represented by four fractions of equal mass, i.e., equal value of Eq. (3) for each fraction,

$$
fm_i = \int_{s_0}^{s_1} f_{dis} ds = \int_{s_1}^{s_2} f_{dis} ds = \int_{s_2}^{s_3} f_{dis} ds = \int_{s_3}^{\infty} f_{dis} ds = \frac{1 - M_6}{4}
$$
 (7)

Therefore, once the s_0 , s_1 , s_2 , and s_3 values have been found, Eq. (4) is again used to calculate the molar mass for each one of the fractions.

It may also be convenient not to include the full analytical description of the light fraction but to lump some of the light components together particularly those with low concentrations. Thus, in this work some of the low concentration light compounds have been lumped together so that the total amount of representative compounds for a given fluid totals 10.

After the C_{7+} is split into a number of representative pseudo-components, the next problem is the assignment of the scaling parameters that are required by the EOS to be used in the fluid phase behavior representation. For the case of a cubic EOS these parameters are the critical temperature T_c , the critical pressure P_c , and the acentric factor ω , for each one of the pseudo-components and lumped fractions. The estimation of EOS scaling parameters has been the subject of a long and extensive discussion in the literature, and a recent revision of the most commonly used correlations is also included in Chap. 5 of Ref. 11. The scaling parameters correlation method used in this work is extremely simple but, in spite of this, it appears to deliver adequate phase behavior performance. The method used in this work, for the light lumped compound groups and heavy fractions scaling parameters estimation, is based on empirical correlations derived from the properties of normal alkanes. The empirical equations are

$$
T_{c,i} = -423.587 + 210.152 \ln(MW_i)
$$
 (8)

$$
P_{c,i} = f_c \exp(9.67283 - 4.05288 \, M \, W_i^{0.1}) \tag{9}
$$

$$
\omega_i = \exp\left(8.50471 - \frac{15.1665}{MW_i^{0.1}}\right) \tag{10}
$$

Here, the molar mass MW_i of the pseudo-component *i* is given in g · mol⁻¹ to obtain the corresponding $T_{c,i}$ in K, $P_{c,i}$ in bar, and acentric factor ω_i . In Eq. (9), f_c represents a perturbation factor away from the $f_c = 1$ value that corresponds to the fit of *n*-alkanes. This perturbation factor is iteratively modified until the fluid saturation pressure is matched this is a key step in the estimation of the scaling parameters. It should also be pointed out that for the well-defined light compounds (i.e., methane, ethane, etc.) tabulated values are used. Also, for the light lumped compounds Eqs. (8) through (10) may also be used to estimate the corresponding scaling parameters using $f_c = 1$ —this gives reasonable results provided the lumped compounds concentration is low.

In addition to matching the saturation pressure, a Péneloux volume translation [12] proposed in Ref. 8 is used in conjunction with this procedure. The volume correction gives accurate density modeling results and consists of shifting the volume from the volume v obtained with the EOS to a translated volume v' ,

$$
v' = v - c \tag{11}
$$

where, in the case of reservoir fluids, c is estimated with the following mixing rule:

$$
c = K_{\mathbf{v}} \sum_{h.f.r.} x_i M W_i \tag{12}
$$

In Eq. (12), the summation only includes the heavy fraction (h, fr) . pseudo-components corresponding to C_{7+} and K_v represents a characteristic volume constant for the fluid. The constant K_v is obtained by tuning against high-pressure density data measured at and above the saturation pressure.

3. OIL VISCOSITY CHARACTERIZATION OF NORMAL OILS

A detailed description of the viscosity modeling procedure for normal crude oils using the f -*theory* models is readily available [3], and only a brief summary will be given here. The application of the f -*theory* to viscosity modeling of crude oils consists of applying the one-parameter f *theory* models to previously characterized reservoir fluids. In the friction theory the total viscosity η is separated into a dilute gas viscosity term η_0 and a residual friction term η_f ,

$$
\eta = \eta_0 + \eta_f \tag{13}
$$

The dilute gas viscosity η_0 is defined as the viscosity at the zero density limit and, for systems such as reservoir fluids, it can be accurately estimated with the empirical model of Chung et al. [13]. The residual term η_f is related to friction concepts of classical mechanics and can be approximated by

$$
\eta_{\rm f} = \kappa_{\rm r} p_{\rm r} + \kappa_{\rm a} p_{\rm a} + \kappa_{\rm rr} p_{\rm r}^2 \tag{14}
$$

where p_a and p_r are the van der Waals attractive and repulsive pressure contributions in the mixture. These contributions can be estimated from simple cubic EOS, such as the SRK EOS or the PR EOS, among others. In the case of alkanes and other nonpolar fluids, including reservoir fluids, generalized models depending on one characteristic fluid viscosity scaling parameter have been further developed [6, 7]. The formulation of the oneparameter general models is as follows:

$$
\eta_{\rm f} = \eta_{\rm c} \left(\hat{\kappa}_{\rm r} \frac{p_{\rm r}}{P_{\rm c}} + \hat{\kappa}_{\rm a} \frac{p_{\rm a}}{P_{\rm c}} + \hat{\kappa}_{\rm rr} \frac{p_{\rm r}^2}{P_{\rm c}^2} \right) \tag{15}
$$

where η_c is the characteristic fluid viscosity scaling parameter. The $\hat{\kappa}_r$, k_a , and k_{rr} parameters are only dependent on the reduced temperature and have been parameterized into universal constants that are related to a specific EOS. For the case of mixtures, the value of the mixture friction coefficients is predicted using the mixing rules suggested in Ref. [6]. For the EOS, in principle, the mixing rules that best describe the fluid phase behavior should be used.

For crude oils, once the fluid has been properly characterized, the determination of the viscosity scaling parameters is similar to that for the $p\gamma T$ scaling parameters. That is, for well-defined light compounds (i.e. methane, ethane, etc.), reported tabulated values [6] are used. For the case where some light compounds (up to C_6) have been lumped together, the following modified Uyehara–Watson correlation [3, 14] may be used:

$$
\eta_{c,i} = 7.94830 \ 10^{-4} \frac{\sqrt{MW_i} P c_i^{2/3}}{T c_i^{1/6}}
$$
(16)

where the units in Eq. (16) are MW in g · mol⁻¹, P_c in bars, and T_c in K to obtain η_c in mPa \cdot s. For the pseudo-components that correspond to the C_{7+} fraction, Eq. (16) is relaxed by substituting the model constant for an adjustable common parameter K_c that can be taken as a viscosity characterization parameter for all of the pseudo-components in the heavy fraction, i.e.,

$$
\eta_{c,i} = Kc \frac{\sqrt{MW_i} P_{c_i}^{2/3}}{T_{c_i}^{1/6}}
$$
\n(17)

Furthermore, it has been shown that substitution of Eqs. (16) and (17) into the one-parameter friction theory models results in a simple linear equation [3] of the form,

$$
\eta = \eta_I + K_c \eta_{II} \tag{18}
$$

where η_I and η_{II} are well defined numbers [3]. Clearly, although to solve for K_c one viscosity point is enough, it has also been found that best results are obtained if K_c is estimated by an optimization procedure using viscosity data from the single-phase region above the fluid saturation pressure.

The approach summarized in this section has been extensively shown to be able to provide accurate viscosity modeling results for reservoir fluids with MW up to around $200 \text{ g} \cdot \text{mol}^{-1}$ [3,8,9,15]. However, as a reservoir fluid becomes heavier, cubic EOSs show systematic compressibility-related deterioration in the quality of the results. To illustrate this, three fluids, a normal one (Oil 1), a slightly heavier one (Oil 2), and a heavy one (Oil 3) have been chosen. The main properties for these three oils are summarized in Table I, along with three other heavy oils, after having carried out a 10-component fluid characterization as described in Section 2 of this work, that is, after lumping some of the light fractions, carrying out the mass characterization procedure for the C_{7+} fractions into four heavy

Fig. 1. Viscosity modeling results for the viscosity characterization constant K_c optimized with data above the saturation pressure. Model predictions: solid lines; experimental data: points.

pseudo-components, and tuning for the saturation pressure using the PR EOS. For the PR EOS the regular van der Waals mixing rules (linear in b and quadratic in a) have been used with the following binary parameters: 0.02 for N₂-C₁; 0.06 for N₂-(C₂-C₃); 0.08 for N₂-C_{i>3}; 0.12 for $CO₂-C₁$; 0.15 for $CO₂-C_{i>1}$; 0 for all hydrocarbon–hydrocarbon interactions $(C_i$ and F_i compound groups). As Fig. 1 shows, the viscosity modeling results, after tuning K_c with data above the saturation pressure, are qualitatively and quantitatively good for Oil 1 and, to some extent also for Oil 2, but not for Oil 3. In the case of Oil 3, it can be seen that the PR EOS based f -*theory* model fails to correctly match the viscosity vs. pressure slope. This problem is consistently found in heavy fluids, and it is thought to be related to the simple mathematical structure of the repulsive term in the cubic EOS.

4. COMPRESSIBILITY CORRECTION FOR HEAVY OILS

4.1. Volume Correction

The friction theory takes advantage of the repulsive–attractive balance built into van der Waals type of EOS. This has been shown to work extremely well for a large variety of fluids for which the EOS gives reasonably good performance. However, for highly dense fluids the simple mathematical form of a van der Waals type of EOS, particularly the repulsive term, is not adequate. Essentially, as illustrated in the Oil 3 example depicted in Fig. 1, the response that the repulsive term gives to the viscosity with the pressure is not as sharp (lower slope) as experimentally observed. Thus, a correction procedure that does not require a mathematical modification of the EOS has been devised. In order to correct the compressibility discrepancy, it has been found that a simple and efficient way to achieve this is by displacing the volume by a factor of ζ ,

$$
\tilde{v} = v - \zeta \tag{19}
$$

and then estimating the viscosity friction contribution using the corrected volume. That is,

$$
\eta_{\rm f} = \kappa_{\rm r} \tilde{p}_{\rm r} + \kappa_{\rm a} \tilde{p}_{\rm a} + \kappa_{\rm rr} \tilde{p}_{\rm r}^2 \tag{20}
$$

where \tilde{p}_r and \tilde{p}_a are the repulsive and attractive pressure contributions estimated at the displaced corrected volume \tilde{v} .

4.2. Mixing Rules

Although the correction described in Eq. (19) is applied in order to correct the viscosity vs. pressure slope in the high-pressure single-phase region, some mixing rules for ζ are necessary for the region below the saturation pressure. Thus, the following mixing rule has been empirically found to provide satisfactory results:

$$
\zeta = K_{\mathbf{z}} \sum_{i} x_i M W_i^{1/3} \tag{21}
$$

where the summation applies only to the pseudo-components of the heavy oil fraction.

5. RESULTS

In spite of the simplicity of the approach presented in the previous section, the introduction of the described corrections results in accurate viscosity modeling behavior above the saturation pressure and satisfactory viscosity predictions below the saturation pressure. Figures 2 and 3 illustrate the modeling results for the heavy fluid previously presented in Fig. 1. In Fig. 2 it can be appreciated that the correction is not required for Oil 1 and it has a slight improvement in the quality of the Oil 2 results. However, as Fig. 3 indicates, the effect of the correction is remarkable for the case of heavy Oil 3.

The performance of this approach for the viscosity modeling of heavy oils is further illustrated for the cases of the Oils 4, 5, and 6 that are

Fig. 2. Viscosity modeling results for Oils 1 and 2. Corrected model predictions for Oil 2: solid line; original model predictions: dashed lines; experimental data: points.

Fig. 3. Viscosity modeling results for Oil 3. Corrected model predictions: solid line; original model predictions: dashed line; experimental data: points.

depicted in Fig. 4. Clearly, the modeling results above the saturation pressure are visibly accurate while the predictions below the saturation pressure are within experimental uncertainty. It is worth noting that the uncertainty in the experimental viscosity measurements of heavy oils below the saturation pressure is extremely high. This is particularly affected by the experimental uncertainty in the equilibrium curve determination and the high possibility of solid formation.

Fig. 4. Viscosity modeling results for Oils 4, 5, and 6. Corrected model predictions: solid lines; experimental data: points.

In addition to the viscosity results obtained with the modified f -*theory* approach presented here, the density modeling scheme discussed in Section 2 has also been applied to Oils 1, 5, and 6—density data for Oils 2, 3, and 4 are not available. For Oils 1, 5, and 6, the density modeling results above the saturation pressure and the corresponding density predictions below the saturation pressure are shown in Fig. 5. In all cases, the absolute average deviation (AAD) of the density modeling results is better than 0.5%.

Fig. 5. Density modeling results for Oils 1, 5, and 6. Corrected model predictions: solid lines; experimental data: points.

	Oil 1	$O11$ 2	Oil ₃	$O11$ 4	Oil ₅	O _i 1 ₆
K_v (cm ³ · g ⁻¹) 0.120213 N/A $^aK_c\times 10^4$ $K_{\rm z}$ (cm ³ · g ⁻¹) 0	7.46396	11.0402 1.12254	N/A 7.34572 2.38373	N/A 34.9945 2.12622	-0.164751 85.1331 1.48187	-0.267768 115.416 1.38409

Table II. Viscosity and Density Parameters

 a Units as defined for Eq. (16).

Finally, the viscosity and density parameters corresponding to all of the examples presented in this work are reported in Table II.

6. CONCLUSIONS

In this work, the previously developed f -*theory* viscosity modeling approach for reservoir fluids [3] has been successfully extended to practically the full range of reservoir fluids that are of interest to the oil industry. Altogether, a comprehensive f -*theory* viscosity modeling approach that goes from the straightforward accurate viscosity prediction of light natural gases [2] to the accurate modeling of heavy oils presented in this work, covering practically six orders of magnitude in viscosity, has been accomplished. The comprehensive approach to the viscosity modeling of reservoir fluids that is extended in this work, is built on the same type of simple tools, i.e. cubic EOS, that are commonly used in the oil industry. Therefore, the incorporation of this approach into other more elaborative oil industry tools, such as reservoir simulators, is straightforward. Furthermore, an efficient implementation of the *f*-theory viscosity modeling approach into a compositional reservoir simulator would not represent any substantial additional computational cost, since it can take advantage of already computed properties. However, the prediction capabilities of the f -*theory* model in the simulation of scenarios with substantial compositional changes, such as gas injection or reservoir depletion, represent a clear advantage over other viscosity modeling approaches that are commonly used in the oil industry, such as the Lohrenz-Bray-Clark (LBC) model [16].

In addition to the f -*theory* viscosity modeling, the approach has been completed with an accurate density modeling scheme. This density modeling approach, also presented in this work, completes two of the most important needs in the oil industry: modeling and prediction of reservoir fluids viscosity and density. As long as the presented density modeling method is applied away from critical conditions, as is the case for most

dense reservoir fluids and certainly for heavy oils, this Péneloux-based density modeling approach is capable of providing accurate results. In all cases studied in this and previous work [8, 15], the presented density modeling approach gives an AAD within 1% for the liquid phase of a wide range of reservoir fluids.

ACKNOWLEDGMENT

The authors gratefully acknowledge TotalFinaElf for the financial support provided to this work.

REFERENCES

- 1. S. E. Quiñones-Cisneros, C. K. Zéberg-Mikkelsen, and E. H. Stenby, *Fluid Phase Equilib.* **169**:249 (2000).
- 2. C. K. Zéberg-Mikkelsen, S. E. Quiñones-Cisneros, and E. H. Stenby, *Int. J. Thermophys.* **23**:437 (2002).
- 3. S. E. Quiñones-Cisneros, C. K. Zéberg-Mikkelsen, and E. H. Stenby, Chem. Eng. Sci. **56**:7007 (2001).
- 4. G. S. Soave, *Chem. Eng. Sci.* **27**:1197 (1972).
- 5. D.-Y. Peng and D. B. Robinson, *Ind. Eng. Chem. Fundam*. **15**:59 (1976).
- 6. S. E. Quiñones-Cisneros, C. K. Zéberg-Mikkelsen, and E. H. Stenby, *Fluid Phase Equilib.* **178**:1 (2001).
- 7. S. E. Quiñones-Cisneros, C. K. Zéberg-Mikkelsen, and E. H. Stenby, *General One-Parameter Friction Theory Viscosity Model for the Patel-Teja EOS*, presented at Equifase 2002, Foz de Iguazú, Brazil (2002).
- 8. S. E. Quiñones-Cisneros, A. Dalberg, and E. H. Stenby, *Pet. Sci. Technol.* (in press).
- 9. S. E. Quiñones-Cisneros, Viscosity Modeling and Prediction of Crude Oils, presented at Equifase 2002, Foz de Iguazú, Brazil (2002).
- 10. C. H. Whitson, *Soc. Pet. Eng. J.; Trans. AIME* **275**:683 (1983).
- 11. C. H. Whitson and M. R. Brulé, Phase Behaviour: SPE Henry L. Doherty Series, Vol. 20 (Soc. Petrol. Engs. Inc., Richardson, Texas, 2000).
- 12. A. Péneloux, E. Rauzy, and R. Fréze, *Fluid Phase Equilib.* 8:7 (1982).
- 13. T.-H. Chung, M. Ajlan, L. L. Lee, and K. E. Starling, *Ind. Eng. Chem. Res*. **27**:671 (1988).
- 14. O. A. Uyehara and K. M. Watson, in *National Petroleum News* (1944), p. R.
- 15. S. E. Quiñones-Cisneros, C. K. Zéberg-Mikkelsen, and E. H. Stenby, *Fluid Phase Equilib.* **212**:233 (2003).
- 16. J. Lohrenz, B. G. Bray, and C. R. Clark, *J. Pet. Technol*. **16**:1171 (1964).