THE DYNAMICS OF RESERVOIR FLUIDS AND THEIR SUBSTANTIAL SYSTEMATIC VARIATIONS

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ABSTRACT

A series of reservoirs are examined to assess the state of their contained fluids in particular with respect to the extent of thermodynamic equilibrium in the reservoir. In addition, this paper resolves the long-standing puzzle as to why tar/bitumen deposition is at the crest of the field in some reservoirs and at the oil-water contact in other reservoirs. Substantial systematic fluid variations are found utilizing Downhole Fluid Analysis (DFA) as the enabling technology. This study employs the cubic equation of state (EoS) for gas-liquid analysis and the Flory-Huggins-Zuo EoS and the Yen-Mullins model of asphaltenes for analysis of dissolved solid - solution equilibria of (live) reservoir crude oils. ‘Young’ fluid systems exhibit huge, non-monotonic variations of fluids (and solids), while moderately aged fluid systems exhibit monotonic yet grossly disequilibrium properties and the ‘aged’ reservoirs are fully equilibrated even in massive scale. Nevertheless, these old reservoirs retain significant fluid and organic solid variations as a result of sequential fluid-related processes in geologic time.

The dynamical behaviors of fluids within reservoirs to account for these variations are obtained by linking a simple understanding of petroleum, especially its asphaltene nanocolloidal and thermodynamic properties, with simple concepts from fluid mechanics. In particular, the location of tar deposition in reservoirs is clarified when formed due to asphaltene instability upon a secondary reservoir fluid charge. Tar deposition can be formed upstructure for rapid gas charge as is regularly seen in young reservoirs, or can be formed at the oil-water contact for a slower gas charge as seen in many older reservoirs. Most importantly, the state of the reservoir fluids within the context of geologic time is shown to be tightly coupled to key reservoir concerns for production. Thus, the understanding of the geologic context of the reservoir can be utilized to optimize reservoir evaluation such as for wireline formation testers (WFT). The expanding capabilities of DFA coupled with formidable advances in petroleum science have revealed dramatic systematic variations of reservoir fluids which can be understood in simple ways. These new methods are becoming indispensable for optimization of production.

INTRODUCTION

Reservoir fluids are one of the most complex chemical mixtures known with tens of thousands of distinct chemical components that can be identified in the asphaltene fraction alone.[Mullins, 2010] Reservoir fluids vary from dry natural gas to tar and all fluids in between.[Tissot & Welte, 1984] In addition, reservoir fluids can exhibit variability in individual reservoirs due to many different fluid and reservoir processes.[Tissot & Welte, 1984; Mullins, 2008] The combined complexities of the chemistry and of the geologic processes might appear overwhelming. Fortunately, there are fundamental scientific simplifications and technological advances that unite to create a powerful approach to reservoir evaluation. Implications of this analysis impact a broad array of disciplines from seismic surveys to fluid mechanics, thereby providing stringent tests for possible specific realizations of the reservoir.

In general, reservoir crude oils consist of dissolved (hydrocarbon) gases, liquids and dissolved (or stably suspended) solids, the asphaltenes (cf. Fig. 1). For the gases, there can be nonhydrocarbon components such as CO₂ and H₂S in the mix. For the solids, the asphaltenes, the qualifier ‘suspended’ appears; generally, the asphaltenes are not dissolved in the liquid as a true molecular solution. Instead, the asphaltenes can self-assemble in crude oil and in laboratory solvents to form stable nanocolloidal species, nanoaggregates...
and clusters of nanoaggregates. At micron and bigger length scales, the fluid appears to act as a true molecular solution of asphaltenes; thus, for many purposes, the asphaltenes can be considered “dissolved”.

![Diagram of crude oil components](image)

**Fig. 1.** Depiction of the composition of crude oils consisting of dissolved gases, liquids and dissolved solids. The thermodynamics of the gas-liquid system is treated with the cubic equation of state (EoS). The thermodynamics of the solution-solid system is treated with the Flory-Huggins-Zuo EoS with its reliance on the nanostructures of asphaltenes embodied in the Yen-Mullins Model.

**Thermodynamic Modelling.** For many reservoir fluids, such as condensates, the variation of dissolved gas content is quite large. This variation depends on the normally large compressibility of condensates due to their large fraction of dissolved gas.[Hoier & Whitson, 2001] The compressibility combined with the hydrostatic head pressure of the oil column creates a density gradient. In turn, the density gradient drives a chemical compositional gradient placing low density components like methane towards the top of the column. Consequently, even though crude oils are complex, the predominant fluid variation in a condensate column can largely be captured simply by measuring the gradient in the gas-oil ratio (GOR). Moreover, such a gradient in GOR can be modeled with a cubic EoS. The first cubic EoS, the van der Waals equation was developed in 1873. Refinements to this equation have been developed enabling applicability to crude oil, such as the Peng-Robinson EoS.[Peng & Robinson, 1976]

In contrast to the gas-liquid equilibrium, there had been no treatment of the fluid-solid equilibrium. The origin of this problem was the lack of any clarity regarding asphaltene nanostructures (molecular or colloidal) in crude oil or even in laboratory solvents. After considerable effort, the nanostructures of asphaltenes were resolved and codified in what is now referred to as the Yen-Mullins Model.[Mullins 2010; 2011] With this resolution, the gravity term and other terms could be specified in an EoS to treat asphaltene gradients. The gravity term consists of accounting for the negative buoyancy of asphaltene particles, the Archimedes buoyancy, in the argument of the Boltzmann distribution. Inclusion of this gravity term into a polymer solution theory gave rise to the Flory-Huggins-Zuo (FHZ) EoS to treat asphaltene gradients.[Freed et al, 2010; Zuo et al, 2013] This theory has strong underpinnings, as Professor Paul J. Flory was a Nobel Laureate. The FHZ EoS treats the fluid-solid equilibrium. The term ‘fluid’ is used as opposed to the term ‘liquid’ because the gas content of the liquid phase has an important impact on asphaltene solubility. The term fluid is meant to include liquid and its dissolved gas.

A well-known axiom in solution chemistry is that “like dissolves like”. Sugar dissolves in water because both have oxygen-hydrogen chemical groups. In contrast, oil and water do not dissolve in each other because oil has carbon-hydrogen chemical groups while water, oxygen-hydrogen chemical groups. Gas is a colorless gas (!) while asphaltene is a dark brown solid. They do not ‘like’ each other in a chemical sense. Asphaltenes do not dissolve in high GOR oils. If gas lift is used (at high pressure) for asphaltenic oils, asphaltene flocculation is expected.

Treatment of the oil column with an Equation of State (without other terms) implies thermodynamic equilibrium, the parameter time does not appear in the equations. Indeed, many reservoir crude oils are in thermodynamic equilibrium, thus an EoS treatment is scientifically valid. However, many reservoirs are not equilibrated. For example, reservoirs undergoing current charging are not equilibrated. In this case, an EoS can still be utilized but only in conjunction with a transient, time-dependent term to treat the ongoing process. That is, equilibrium implies time independence without any net fluxes of chemical components or heat. If a flux, for example of a gas charge, is in fact occurring in a reservoir, then it is not in equilibrium. In such cases, a time-dependent term can be included with an EoS to treat the reservoir crude oil. This protocol extends the thermodynamic treatment of equilibrated reservoir crude oils to those reservoir crude oils that are not in equilibrium. Naturally, if a particular process is presumed with its corresponding thermodynamic treatment, it behooves the analyst to find evidence that this presumed process is in fact correct.

One of the best methods to test the applicability of any thermodynamic treatment of reservoir crude oils is to measure fluid compositional variations vertically and
laterally within the reservoir. Moreover, because there are two EoSs (the cubic EoS for gas-liquid and the FHZ EoS for the fluid-solid) for the three different components, then there are two independent measurements providing an even more stringent test of any thermodynamic model of the reservoir crude oils. Essentially, it is readily possible to have a spurious fit to a few data points for example for a very sparse fluid data set. It is much more difficult to have a spurious fit to a relatively rich fluid data set that consists of vertical and lateral fluid gradients. In particular, both the GOR and the asphaltene content are constrained thermodynamically as to how they can vary spatially. Fluid gradient measurements effectively invoke this constraint in the test of a thermodynamic model.

**DFA.** The best method to measure the fluid gradients is to employ Downhole Fluid Analysis (DFA).[Mullins, 2008] Figure 2 shows an image of a wireline formation sampling tool outfitted with two DFA tools. DFA performs fluid measurements with the least possible fluid handling. Performing the DFA measurements within a single well enables detection of very small fluid gradients as all systematic errors cancel. The latest DFA tools are designed to provide similar abilities to detect small gradients across the field. DFA also offers the ability to optimize robust measurement of fluid gradients in real time.

**Universal Workflow.** Reservoir case studies employing DFA measured gradients with thermodynamic analysis based on the cubic EoS and the FHZ EoS have addressed a wide variety of topics. Figure 3 shows a schematic of this protocol along with several of the topics that have been addressed.

**Fig. 2.** Wireline formation sampling tool equipped with two Downhole Fluid Analysis (DFA) tools. DFA is indispensable for reservoir evaluation.[Mullins, 2008] fluid gradients are best measured with DFA. Thermodynamic treatment of these gradients for gas-liquid using the cubic EoS and for solid-solution using the FHZ EoS yields a stringent test of any fluid model.

**Fig. 3.** A schematic of the universal workflow employed to address a myriad of reservoir complexities. Fluid compositional gradients are measured by DFA. The gradients are then analyzed by a proper thermodynamic treatment. The cubic EoS is used for gradients of dissolved gas-liquid and the FHZ EoS is used for gradients of fluid – dissolved solid, the asphaltenes. In all case studies to date, this protocol added significant understanding to the reservoir issues of concern.

Recent reports using this work flow of DFA gradient measurement coupled with thermodynamic analysis includes fault block migration via asphaltene gradients [Dong et al, 2012], connectivity [Pfeiffer et al, 2011], biodegradation, water washing and multiple charging, the origin of heavy oil gradients [Pomerantz et al, 2013], Flow Assurance [Chen et al, 2014], and CO2 [Quayle et al, 2013]. When new science and new technology are linked as is the case here, there is always an explosion of applications.

**CONNECTIVITY**

Evaluation of reservoir connectivity is virtually always of interest. For deepwater and other high tier markets where well costs preclude field development plans with large numbers of wells, reservoir connectivity becomes a crucial concern. Methods to evaluate connectivity in the past have not performed well. Figure 4
demonstrates this fact in a clear manner. An industry study of 28 deepwater Gulf of Mexico reservoirs was performed.[cf references in Chen et al, 2014] Of these reservoirs, 75% were found to underperform in both production rate and in total recovery. Unrecognized compartmentalization was found to be the most important culprit in this underperformance. It is obvious that new methods of connectivity analysis are needed. Moreover, these reservoirs are already in production. For reservoirs currently being evaluated, the economic margins are tighter than in the past; this kind of underperformance is even less acceptable.

Asphaltenes are an important component of what follows both for the FHZ EoS and fluid mechanics considerations as well as the reservoir fluid dynamics that give rise to the observed distributions of fluids and organic solids in reservoirs. The molecular and colloidal nanostructures are given in the Yen-Mullins Model and are shown in Fig. 5. Professor Teh Fu Yen founded modern asphaltene science in the 1960s. This model was introduced and discussed in review articles.[Mullins, 2010; 2011]

Figure 6 shows case studies of five different reservoirs where the asphaltenes were equilibrated as shown by FHZ EoS analysis and connectivity was established in production, the ultimate arbiter.[Zuo et al, 2013] These case studies span condensates to heavy oils proving a broad range of applicability of the FHZ EoS. There is no other EoS of asphaltenes that has been shown to work over this range. Figure 6 also shows the relevant asphaltene moieties from the Yen-Mullins Model (cf. Fig. 5) for each of the case studies. For the condensate, the very light fluid has virtually no asphaltene, it is the heavy resins that established the heavy end gradient. n.b. Connectivity does not mean the asphaltenes are equilibrated, many reservoir are undergoing current processes which preclude asphaltene equilibration. For dynamic reservoir processes, the FHZ EoS plus a dynamic term accounting for this reservoir fluid process can then be used to analyze DFA gradients and assess connectivity.

In many case studies, the evaluation of the light ends by the cubic EoS did not reveal the same physics and reservoir understanding as the evaluation of the asphaltenes by the FHZ EoS. For example, in a reservoir study (Fig. 6, middle plot) [Dong et al, 2012], the asphaltene gradients indicated connectivity which was proven in production, while the light end analysis was ambiguous. Another case study involved a low GOR black oil (Fig. 6, 2nd from right).[Betancourt et al, 2007; Mullins et al 2007] Low GOR crude oils have
largely homogeneous GORs, thus in these cases, GOR cannot be used effectively to check for thermodynamic equilibrium. In this low GOR case study, asphaltene equilibration was used to predict connectivity in each of two stack sands, results later born out in production.[Creek et al, 2010]

Fig. 6. DFA studies establish likely connectivity if the asphaltenes are equilibrated.[Zuo et al, 2013] These cases span condensates to heavy oil. The green curve is the FHZ EoS fit using the embedded asphaltene species from the Yen-Mullins Model. For reservoirs with disequilibrium of asphaltenes, the FHZ EoS analysis includes other theoretic terms to account for disequilibrium in connectivity studies.

In another reservoir study (Fig. 6, 2nd from left), the reservoir had two separate gas caps in a light oil reservoir.[Gisolf et al, 2009] The GOR analysis suggested discontinuity because the two gas caps had separate gas-oil contacts (GOCs) that differed by 20 meters true vertical depth (TVD). In contrast, the asphaltenes indicated connectivity by exhibiting a continuous and equilibrated gradient (by FHZ EoS) across the field.[Gisolf et al, 2009] The asphaltene analysis was proven correct in production. The salient feature is that the light ends and the asphaltenes can be controlled by different physical processes in the reservoir. It is essential to understand these distinctions to interpret important reservoir properties such as connectivity. For example, in this reservoir, there was a late gas charge. It would be almost impossible for the two separate gas caps to receive proportional quantities of gas to keep the two GOCs the same. The cubic EoS correctly predicted disequilibrium which might be construed as disconnected reservoirs. Even though there was a gas charge into the reservoir, the GOR was not changed dramatically (there were gas caps), so the asphaltene distribution was also not significantly altered by the gas charge. Thus, the equilibrated asphaltenes were used to predict connectivity.[Gisolf et al, 2009]

GAS CHARGE INTO OIL RESERVOIRS

A late charge of gas or light hydrocarbon into reservoirs is rather common. Figure 7 shows a gas chimney, this one in the North Sea, that is large in comparison to reservoirs shown.[Granli et al, 1999] Gas charging into a reservoir can have a dramatic effect on the oil if the reservoir pressure is sufficient to put this gas in solution. Specifically, asphaltenes can be destabilized by an increase in solution gas. The fate of the destabilized asphaltenes depends on the specifics of gas entry rates, thermodynamic considerations and asphaltene nanoscience.

There is a substantial difference between the outcome for asphaltene destabilization depending on whether the solution gas increases rapidly or slowly. For example, an EOR process of miscible gas flood amounts to extremely rapid gas addition in comparison to geologic time. In such cases, the GOC (or the transition zone between the gas and oil) sweeps past points in the reservoir. The asphaltene in any residual oil is destabilized to the point of phase instability and deposits locally.[Pedersen et al, 2012] There is no time for the asphaltenes to ‘get out of the way’. In contrast, a very slow gas charge will give rise to a diffusive front of solution gas increase. That is, the gas can diffuse into the oil. In this case, the asphaltenes have ample time to migrate. Such a slow gas charge can result from biogenic gas.

We compare the rate of a GOC sweep vs. the rate of a diffusive front for ‘typical’ reservoir scenarios.
Consider a tilted sheet sandstone reservoir with a 10° dip angle and with a TVD height of 100 meters (and reservoir length of 576 meters), sufficient for our considerations. Typical reservoir charge times are 1 to 10 million years. If we take 3.3 million years as our charge time, then we get a GOC vertical displacement of ~30 meters/ million years.

Now consider the rate of a methane diffusive front in this reservoir. The one dimensional diffusion equation gives $2Dt = x^2$ where $D$ is the diffusion constant, $t$ is time, and $x$ is distance along our reservoir formation. A reasonable diffusion constant of methane is $10^6$ cm$^2$/sec. To diffuse 15 meters vertical would take about 1 million years. This is almost the same number obtained by GOC displacement above. The conclusion is that geologic gas charging processes can have either the contact displacement dominate or the diffusive front dominate. Faster gas charge would have the contact displacement dominate (as it does in miscible flood EOR) while slower gas charge would enable the diffusive front to dominate.

An important point in these considerations is length scale. We have used ~20 meters as our characteristic length scale of comparison. GOC displacement is essentially linear in time while for diffusion, the square of the displacement $x$ is proportional to time. Because these two processes scale differently, a characteristic length is required for comparison. We have used in the above a characteristic length of ~ 20 meters vertical. If we chose 2 centimeters as a characteristic length, then diffusion would always be faster than displacement. If we chose 2 kilometers as a characteristic length, then diffusion would always be slower than contact displacement. The justification for ~ 20 meters as our characteristic length is due to the requirement that a sufficient volume of fluid, not much more and not much less, be considered for our reservoir fluids processes (the Goldilocks rule). 20 meters vertical (and ~100 meters measured depth) provides such a volume of fluid to be meaningful for the following reservoir considerations.

From the mass perspective, similar considerations apply to avalanches. Considering too small a length scale, thus too little mass, would give rise to tiny disturbances that do not produce avalanches. Considering only a very large a length scale, thus considering only very large masses, does not capture important avalanches.

**SLOW GAS CHARGE** Figure 8 shows a schematic is helpful to describe the process of slow gas charge to the reservoir. Some of the scientific details of this process are still being worked out (such as the cluster diffusion rates in a chemical potential gradient); nevertheless, there is ample evidence for the major features of the proposed mechanism. Four time snapshots of the reservoir are shown with time going from left to right. The original reservoir prior to a gas charge is on the left. Next, some gas charges into this reservoir with sufficient reservoir pressure that the solution gas increases. The gas is expected to go to the top of the reservoir without mixing.[Stainforth, 2004] In reality, the gas might mix with local oil creating a high GOR light oil that rises to the top of the reservoir; the considerations remain the same. The gas then diffuses down into the oil column increasing solution gas and expelling asphaltene. As gas diffuses into the oil, the gas cap diminishes keeping the rate of contact displacement minimal. With slow gas addition, the asphaltenes are destabilized from the small nanoaggregates to the larger asphaltene clusters. Such asphaltene cluster formation is known in many laboratory studies [Mullins, 2010; Eyssautier et al, 2012; Majumdar et al, 2013] and in the reservoir,[Mishra et al, 2012; Seifert et al, 2012; Pastor et al, 2012] The clusters diffusively fall in the gravitational and chemical potential GOR gradient concentrating at the base of the diffusive front thereby increasing the mass density there. The gas content also increases thus decreasing density.

Because the asphaltenes are swept out of the entire column above, the asphaltene concentration increase is integral. The increase in gas at the base of the diffusive front is only differential. Thus, the net density change is an increase thereby giving rise to a density inversion, and convection.

If asphaltenes could only destabilize to flocs and not to a stable nanocolloidal cluster, then the flocs would not be able to migrate at all in the porous formation. The flocs would fall within the pore space, land on a sand grain and remain there for geologic time. Thermal energy would not be able to lift the micron-sized flocs so asphaltene diffusion would cease. Thus, tar mats would form at the GOC. Instead, mild instability of asphaltenes causes formation of asphaltene clusters from nanoaggregates (cf. Fig. 5). The clusters are small enough (5nm) that they can readily diffuse within the porous medium. Density inversions form as depicted in Fig. 8, thus convection occurs.

The steady state velocity $V$ of the convective current is given by:
where $\Delta \rho$ is the density inversion, $g$ is earth’s gravitational acceleration, $k$ is permeability, $\theta$ is the dip angle, $\phi$ is the porosity and $\mu$ is the viscosity. For the case of 100 mD permeability, 20% porosity, 1 cP viscosity and 10° dip angle, Table 1 gives convective flow velocities for different density inversions. Even for moderate density inversions, the gravity currents are fast and can easily span reservoir dimensions. All petroleum system models charge reservoirs via gravity currents (oil or gas vs. water); gravity currents are not a new concept in reservoirs.

Table 1. Gravity current velocities in meters per million years (cf. Eq. 1) for typical reservoir parameters vs. the magnitude of the density inversion.

<table>
<thead>
<tr>
<th>$\Delta \rho$ (g/cc)</th>
<th>$V$ (m/MMY)</th>
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<tbody>
<tr>
<td>1e-3</td>
<td>27000</td>
</tr>
<tr>
<td>1e-4</td>
<td>2700</td>
</tr>
<tr>
<td>1e-5</td>
<td>270</td>
</tr>
<tr>
<td>1e-6</td>
<td>27</td>
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<tr>
<td>7.4e-7</td>
<td>20</td>
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<tr>
<td>1e-7</td>
<td>2.7</td>
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To the left in Fig. 9, the well-known series of dead oil samples are shown. The asphaltenes have been expelled from the oils towards the top of the column, little color remains. This expulsion is in accord with the FHZ EoS [Zuo et al., 2011] utilizing the local GOR. That is, the GOR is grossly out of equilibrium in the oil column. Nevertheless, the asphaltene concentration is locally equilibrated at each height in the reservoir in accord with its local GOR.

Fig. 9 shows a reservoir that is in the process depicted in Fig. 8.[Zuo et al., 2011] A late gas charge is entering the reservoir. The source of the gas is biogenic, its flux is rather small, thus, the gas charge is categorized as slow meaning that the diffusion of gas into the oil column (in the characteristic length of ~20 meters) is fast in comparison to the GOC displacement.

Figure 9 depicts a reservoir that is in the process of gas diffusion from the top along with asphaltene gravity currents giving rise to asphaltene accumulation at the base of the reservoir.[Zuo et al., 2011] Towards the top of the reservoir, there are huge, disequilibrium gradients of GOR and saturation pressure (Psat) due to gas diffusion into the column from the top. Towards the base of the column, there are only very small GOR and Psat gradients; the diffusive gas front has not reached this part of the reservoir. At the top of the reservoir, the asphaltenes have been expelled (visually seen on the left) due to the increase in solution gas. There is a large gradient of asphaltenes at the base of the reservoir consistent with a quasi-gravitational equilibrium of asphaltene clusters. Gravity currents from the top of the reservoir are fast and can easily span reservoir dimensions. All petroleum system models charge reservoirs via gravity currents (oil or gas vs. water); gravity currents are not a new concept in reservoirs.

![Figure 8](image1.png)

Fig. 8. Depiction of time evolution (left to right) of a slow gas charge into an original oil reservoir (leftmost). Gas goes to the top of the oil column and diffuses into the oil column. (2nd from left). As diffusion continues, asphaltenes accumulate towards the base of the diffusive front as clusters giving rise to a density inversion, with convection immediately occurring (3rd from right). As gas continues to charge, asphaltene clusters are pumped by convection (gravity currents) to the base of the reservoir (rightmost). With sufficient asphaltene accumulation at the base, a tar mat forms.

![Figure 9](image2.png)

Fig. 9. A reservoir that is in the process of diffusive gas entry from the top (cf. Fig. 8).[Zuo et al., 2011] The gas diffusion gives rise to large disequilibrium gradients of GOR and saturation pressure (right) towards the top of the reservoir. The expelled asphaltenes are pumped via convective gravity currents to the base of the reservoir yielding large gravitational gradients of asphaltene clusters (center).
reservoir are pumping asphaltenes to the base of the reservoir in the form of clusters whereupon they diffusively set up a gravitational gradient. Biodegradation is also contributing to the asphaltenes accumulation at the base of the column but is thought to be secondary here.

Figure 10 depicts a reservoir, a four-way sealing anticline, with a crude oil that has come to thermodynamic equilibrium as shown by analysis utilizing the FHZ EoS. [Seifert et al, 2012] As is often the case, the cubic EoS analysis is not useful for this reservoir while the FHZ EoS is extremely useful. The reason is simple; low GOR black oils and heavy oils are not defined primarily by their dissolved gas content. Instead, these crude oils are defined by their asphaltenes content. For example, for such oils, their viscosity is often key for determining economic viability or projects and even in assessing important constraints such as placement of water injectors. And for black oils and heavy oils, viscosity is exponentially dependent on asphaltenes content. The FHZ EoS is the only EoS that has been proven to treat heavy oil gradients, due in part to its incorporation of asphaltenes clusters of the Yen-Mullins Model.

In the geologic past, this reservoir (Fig. 10) had experienced a slow, long instability of asphaltenes most likely at the crest and most likely due to a light hydrocarbon charge. The asphaltenes did not deposit locally, upstructure. Instead, the asphaltenes were pumped convectively to the base of the column in asphaltenes cluster gravity currents in the manner depicted in Fig. 8. As seen in Fig. 10, the asphaltenes clusters established a gravitational gradient over the entire 100 kilometer rim of the reservoir. [Seifert et al, 2012] The instability event continued pumping sufficient asphaltenes to the base that the asphaltenes concentration then exceeded the solvency of the crude oil for asphaltenes. For this crude oil with a relatively high aromatic fraction, the solvency limit of asphaltenes is ~30% asphaltene, a rather large fraction. As asphaltenes continued to be pumped to the base of the reservoir, and as gravity continued to concentrate the clusters at the base of the reservoir, a tar mat formed. The tar mat grew to approximately 10 meters in height. The final heavy oil column is about 50 meters in height. And the heavy oil is shown in Fig. 10 to be equilibrated, matching the FHZ Eos predictions almost exactly around the entire 100 kilometer rim of the oil field.

Specifically biodegradation (which occurs at the oil-water contact) could not be responsible for the gradients seen in this reservoir depicted in Fig. 10. Biodegradation increases the asphaltenes content in the crude oil as the microbes preferentially consume other
components. The microbes first consume n-alkanes and other simple saturate compounds. Upon disappearance of the saturates, the microbes will consume simple alkyl aromatics as given in the Peters-Moldowan scale of biodegradation (cf. Fig. 11B). In this manner, the asphaltenes become concentrated. However, in Fig. 10, it is seen that the asphaltene fraction goes from ~3% to ~30%. For this increase to occur via biodegradation, the microbes would have to consume 90% of the crude oil, which is extremely unlikely. Figure 11 shows the SARA (saturates, aromatics, resins, asphaltenes) of the heavy crude oil samples of Fig. 10. The saturate fraction is seen to decrease from ~50% to ~30% as the asphaltenes increase from 3% to 30%. There is no way the microbes could consume 90% of the crude oil, thereby increasing the asphaltene fraction tenfold, while consuming less than ½ the saturates.

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Figure 12A, shows a comparable analysis of the tar samples obtained from core plugs taken from whole core in two tar wells. The asphaltene content in these tar samples ranges from ~37% to ~54%. As with the heavy oil samples above this tar mat, the reduction of the saturates is rather modest from 20% to 15%. Fig. 12B, also shows that this 5% absolute decline of the saturates is comparable to the 5% decline of the aromatics and even to the 5% absolute decline of the resins. Clearly, the large increase in asphaltene content does not result from biodegradation; the microbes would consume all saturates before consuming ½ the resins.

![Graph A](image)

![Graph B](image)

**Fig. 11.** A) SARA fractions of the heavy oil samples plotted in Fig. 10. The asphaltene fraction increases tenfold while the saturates decline by less than a factor of two; biodegradation cannot account for its increase in asphaltene content. B) The Peters-Moldowan Scale of biodegradation showing that the microbes consume simple alkanes prior to consuming cyclic alkanes and other complex components.

**Fig. 12.** A) the SARA fractions from extracted organics in core plugs take from whole core in tar zones. The comparable decline of saturates, aromatics and resins as asphaltenes increase shows that biodegradation cannot be the origin of this tar formation. B) Ratios of SARA fractions indicate that this tar appears to be asphaltene added to (invariant) oil, consistent with the gravity current model of tar mat formation.
Comparison of Figures 11 and 12 seem to indicate that the tar zone has more aromatics than saturates while the heavy oil has more saturates than aromatics. The implication is that the high asphaltene materials preferentially associate with aromatics over saturates. However, the analyses of the tar samples were performed at very different times by different laboratories. Processing of whole core and core plug samples is very different than processing of wireline formation testing samples. And no such trends are seen within the oils alone, or within the tar samples alone. Consequently, while this difference in saturate to aromatic fractions is intriguing, it can only be viewed as suggestive at best.

Fig. 10 shows that the asphaltene clusters have equilibrated matching the FHZ EoS; the GOR is low, thus the only significant term of the FHZ EoS is the gravity term given in Eq. 2. The only adjustable parameter is the asphaltene cluster size. The fit gives 5.1 nm, whereas the Yen-Mullins Model depicts a nominal cluster size of 5.0 nm.[Seifert et al, 2012] For nanoscience, these are identical numbers.

$$\frac{A_h}{A_o} = \exp \left\{ - \frac{V \Delta \rho g h}{kT} \right\}$$  \hspace{1cm} 2.

where $A_h$ is the asphaltene concentration at height $h$, $A_o$ is the asphaltene concentration at the reference height, $V$ is the volume of the particular asphaltene species of interest (cf. Fig. 5), $\Delta \rho$ is the density contrast between asphaltene and the liquid phase of the crude oil, $g$ is earth’s gravitational acceleration, $k$ is Boltzmann’s constant and $T$ is temperature. $V \Delta \rho g$ is simply Archimedes Buoyancy. Eq. 2 is the gravitation Boltzmann distribution $\exp\{-\Delta E/kT\}$. Gravity pulls asphaltenes towards the base of the column while thermal energy lifts the asphaltenes above this base. The Boltzmann distribution gives the balance of these two opposing factors. In Eq. 2, by replacing $V \Delta \rho$ with $m$ for the molecular mass of the atmospheric gases, the barometric equation is obtained; this equation gives the atmospheric pressure gradient of planet earth. Here, the air molecules are suspended in a vacuum, not in a crude oil, so Archimedes buoyancy is replace by mass.

There have been two primary explanations for asphaltene gradients in heavy oil: the “atmospheric-pressure-gradient model” (Eq. 2, consistent with the FHZ EoS and the Yen-Mullins model) and the “sand-storm model” (cf. Fig. 13). The atmospheric-pressure-gradient model stipulates that all nitrogen and oxygen molecules in the atmosphere are the same and the reason that atmospheric pressure is greater at sea level than at the summit of Mt. Everest is that there are more air molecules at sea level than at the summit of Mt. Everest. Likewise, in accordance with the Yen-Mullins model, there is a fixed size of asphaltene clusters, and at the base of the heavy oil column depicted in Fig. 10 there are more clusters as given by Eq. 2.

Fig. 13. Left: Atmospheric pressure versus height; the volumetric density of $N_2$ and $O_2$ molecules declines with height in accord with the barometric equation similar to Eq. 2, thereby giving reduced pressure with height. Equilibrated heavy oil gradients are closely related with a decrease of asphaltene cluster concentration with height. Right: sand storm where larger, denser sand grains are closer to the ground. This model does not mimic heavy oil gradients.

The ‘sand-storm’ model stipulates that in a sand storm, larger, denser sand grains are close to the ground, intermediate sized sand grains are at heights of several meters, and clays are at tens of meters and higher. It is well known that in sand storms, the scouring action is most severe close to the ground. There can be mineral and size sorting with height. For the sand-storm model, the particle sizes and/or mineral composition are changing giving rise to a gradient of suspended solids in the air column, there is not a fixed sand grain size.

This ‘sand-storm model’ has never been shown to apply to asphaltenes. Quite the opposite, a growing number of studies shows this model does not apply. For oil columns with very large gradients of asphaltenes, the chemistry of the asphaltenes has been shown to be invariant. For example, the sulfur chemistry of asphaltenes in the column of Fig. 10 has been shown to be invariant.[Pomerantz et al, 2013] Sulfur is chemically reactive (consider H$_2$S) and a good candidate to show chemical variability if any existed.
In addition, for asphaltenes, no other stable colloidal particle has been detected in laboratory experiments beyond what is shown in Fig. 5; nevertheless, we consider the impact of the possibility of another asphaltene colloidal particle. Suppose a ‘cluster of clusters’ existed with an aggregation number of 10. In the reservoir the half-height for asphaltene clusters, the height at which they drop to ½ the concentration (similar to half-life) is 20 meters (cf. Eq. 2). For a supposed cluster of clusters with an aggregation number of 10, the half-height would be 2 meters. Consequently, they would be undetectable except at the very base of the oil column. Nevertheless, it is desirable to measure asphaltene aggregation directly from samples in a highly graded column.

To differentiate between the atmospheric-pressure-gradient model and the sand-storm model, it is desirable to check if the asphaltene nanostructures are invariant for the highly graded oil column in Fig. 10. Invariant asphaltene nanoaggregate structures support the atmospheric-pressure-gradient model and refute the sand-storm model. Several asphaltene samples from the oil column depicted in Fig. 10 were analyzed. Fig. 14 shows results from laser mass spectroscopy (surface assisted laser desorption ionization, SALDI-MS) that the nanoaggregate aggregation number is about 6 for all asphaltenes from the top to bottom of the column depicted in Fig. 10.[Wu et al., 2014] The aggregation number of 7 is depicted in Fig. 5, showing excellent agreement with this new data. Previous work shows that the molecular weight is the same for all these samples.[Pomerantz et al., 2013] The results confirm the FHZ EoS; that is, the ‘atmospheric-pressure-gradient’ model (Eq. 2) is applicable to asphaltene gradients, the ‘sand-storm model’ is not applicable.

Fig. 14. Laser mass spectrometer methods have been utilized to measure the mass (or m/z with z=1) of asphaltene nanoaggregates.[Wu et al., 2014] These ‘blue’ samples were taken at various locations in the heavy oil column depicted in Fig. 10 and the ‘red’ samples in a separate horizon in the same field. First, the aggregation number, 6, closely matches the Yen-Mullins Model. Second, all nanoaggregates are found to be the same supporting the ‘atmospheric-pressure-gradient model’ encompassed by the FHZ EoS.

RAPID GAS CHARGE When the gas charge to the reservoir is rapid a very different scenario emerges and very different production concerns are encountered. Figure 15 shows a schematic of a rapid gas charge into the reservoir. In this case, the gas-oil contact displaces much faster than diffusion, so diffusion is ignored. There is a transition zone depicted in going from the undisturbed original oil vs. the newly charged gas.[Pedersen et al., 2012] In the transition zone, the quality of the solvent rapidly becomes poor for asphaltenes as the solution gas increases quickly. Any residual crude oil in the transition zone deposits asphaltenes locally.
Fig. 15. Depiction of time evolution (left to right) of a rapid gas charge into an original oil reservoir (left most).[Pedersen et al, 2012] Gas goes to the top of the oil column and the gas-oil contact (GOC) rapidly sweeps down (middle). A transition zone develops between the undisturbed original oil and the newly charged gas. Any residual oil in this zone experiences a rapid increase of solution gas and expels its asphaltenes. No density currents occur. As more gas charges into the reservoir, the GOC contact continues to displace downward.

As discussed previously, in a high pressure setting, the newly charged gas might mix with crude oil in the migration path making the newly charged fluid more similar to a high GOR light oil as opposed to a pure gas phase. Nevertheless, the scenario depicted in Fig. 15 applies.

Figure 16 shows data from two Pliocene fields that both exhibit upstructure bitumen deposition. Both wells are near the crest of their respective fields. In whole core in both wells, the bitumen is found above shale breaks coating up to ½ of the producing interval.[Dumont et al, 2012] In one well, a one foot interval in the bitumen zone was perforated, then straddled with the Wireline Formation Tester Dual Packers, then produced. A light oil of high GOR and low asphaltene content flowed proving the bitumen did not seal the zone.[Dumont et al, 2012] That is, the bitumen only coated the grains and represented a small fraction of the total porosity. Because ~½ the producing interval has bitumen deposition, and the original oil has only a small fraction of asphaltene, there was not sufficient asphaltene to seal ½ of the producing interval. Consequently, it was presumed, then confirmed, that these bitumen zones are permeable in contrast to standard tar mats.

The data in Fig. 16 is comparable to the schematic in Fig. 15. Density-neutron crossover indicates a higher GOR towards the top of the shale-sand sequence. Large influx of gas is known to be occurring in both fields, and nearby sands are gas filled. Two hydrocarbon phases are present in the formation, a light oil of ~1000 scf/bbl with <1% asphaltene and a bitumen of ~30% asphaltene. One feature of the bitumen deposition is that the asphaltenes did descend somewhat in each producing interval before undergoing phase separation to form the bitumen. This is likely related to the GOR gradient evident in the density-neutron cross over. As expected, both fields are known to be at or near their asphaltene onset pressures. The asphaltenes are chemically polydisperse; their stability in oil spans a range of conditions.[Buckley et al, 2007] After the solution GOR increased due to late gas charge, then much of the asphaltenes (and some resins) phase separated. The remaining heavy ends in the reservoir crude oil are on the edge of stability and can phase separate with a pressure reduction giving rise to Flow Assurance problems.
The bitumen deposit can flow (or ooze) when the light oil in the formation is produced. Another reservoir with similar circumstances gave rise to bitumen coating on the production tubing in a well test.

RESERVOIR FLUID DYNAMICS AND PRODUCTION CONCERNS

Very different production concerns are found to depend on the nature and timing of the reservoir fluid dynamics and apply to specific reservoirs. In order to interpret the DFA measured fluid distributions, it is important to understand the context of those measurements. For relatively static fluid systems of sufficient age, Fig. 6 establishes that asphaltene equilibrium can be used as a strong indicator of reservoir connectivity. As reviewed above, other reservoirs exhibit dynamic fluid columns that are grossly out of equilibrium. Nevertheless, thermodynamic modeling can proceed using the cubic EoS and the FHZ EoS with transient terms as long as the dynamic process is understood. To understand dynamics, it is of immediate concern to understand the time frame of the various fluid processes.

The reservoir fluid dynamics are generally on the order of millions of years, not hundreds of thousands of years and not tens of millions of years. Reservoir charging typically takes a few million years, diffusive processes that initiate gravity currents millions of years, vertical (or local) diffusive equilibration of accumulated asphaltene clusters typically takes millions of years, and gravity currents themselves take millions of years. Some of these processes take place at least partially concurrently. Consequently, if reservoirs are considered relative to this millions-of-years dynamic time frame, systematics should be observed.

Figure 18 shows six reservoirs spanning an enormous range of time from Pliocene to Triassic. Many other reservoirs map into this same dynamic scale with the same production concerns. We use the age of the reservoir as a proxy for the age of the fluid system, while considering the more recent fluid processes as well. DFA reservoir analysis that encompasses a petroleum system perspective, combined with the thermodynamics of reservoir fluids and the fluid mechanics of the reservoir fluid processes and high resolution analytical chemistry is the most comprehensive approach. Nevertheless, significant progress can be made within the heuristic approach presented herein from the different disciplines to help identify possible key production concerns.

Figure 18. A projection along the time line of the reservoir formations. [Mullins et al, 2013] Below the image for each reservoir, production concerns are listed that reflect the age and dynamics of the reservoir. The youngest reservoirs (left) contain fluids that are not even monotonic, thus certainly not equilibrated. Reservoirs with slow, current charging (2nd from left) contain monotonic but disequilibrium fluid gradients. Older reservoirs (middle) contain equilibrated fluids. The oldest reservoirs contain equilibrated fluids but possess important signatures of past fluid processes (two reservoirs on right). To reflect the enormous time span involved, prominent animals living at the time of the formation age are shown to indicate the enormity of the different times.

Some of the younger reservoirs are undergoing current fluid dynamics that continue to alter the fluid distributions in important ways. Each reservoir is shown in Fig. 18 with specific production concerns associated with that reservoir. Pliocene reservoirs are often currently charging, and do not have nearly enough
time to equilibrate. Indeed, the fluids in these reservoirs are often not even spatially monotonic.[Dumont et al, 2012] This has important implications for methods to determine connectivity. Indeed, in such reservoirs a fluid density inversion measured across the field cannot be used to prove compartmentalization. For example, a young reservoir with current charging and with faulting radiating away from a salt keel can yield a density inversion in a vertically baffled, but not compartmentalized field. Older reservoirs with slower current charging can still exhibit disequilibrium but will be likely possess spatially monotonic fluid properties.

The Lower Miocene reservoir without any evident significant secondary fluid process has an equilibrated distribution of asphaltenes. Indeed, the gravity term of the FHZ EoS dominates because this is a low GOR black oil and the nanoaggregates are graded according to Eq. 2.[Betancourt et al, 2007; Mullins et al, 2007] This reservoir plays the role of establishing a lower limit for the minimum time a black oil requires to equilibrate in a relative large field, ~20 million years. This is very consistent with the precepts discussed herein about the time required for the different fluid processes to take place as they lead toward a static, equilibrated system. Indeed, this case study illustrates the importance of this type of DFA reservoir analysis to address connectivity, a problem which has been so vexing to the industry (cf. Fig. 4).

The oldest reservoirs show equilibrated fluids even after significant events subsequent to charging. The Saudi Aramco field had a long, slow instability event, asphaltene gravity currents, heavy oil and tar mat formation and with ultimate equilibration of the heavy oil column. The tar mat and the heavy oil column have significant implications for production. The Triassic reservoir underwent significant CO₂ addition likely during Paleogene magmatic intrusion. The contents of this gas reservoir are equilibrated throughout even above and below a potential shale break.[Quayle et al, 2013] There was ample time for equilibration processes to take place. It would have been better for the Operator if the CO₂ were sequestered naturally at some place in the reservoir. Again, important production concerns are addressed by understanding the state of the reservoir fluids in terms of DFA measurements, reservoir fluid processes and thermodynamics.

CONCLUSIONS

Reservoir fluid distributions result from complex processes and, of course, must comply with governing thermodynamic and fluid mechanic laws. Prior to the advent of Downhole Fluid Analysis (DFA), it was difficult to measure these gradients with accuracy. At present, DFA surveys of fluid gradients are becoming commonplace. Crude oils generally consist of three phases, dissolved gases, liquids and dissolved solids, the asphaltenes. In the recent past, the only thermodynamic treatment for crude oil gradients utilized theory applicable for gas-liquid equilibria only, such as the cubic EoS. The resolution of the nanostructures of asphaltenes embodied in the Yen-Mullins Model allowed the gravity and other terms to become specific. In turn, this enabled the development of the Flory-Huggins-Zuo (FHZ) EoS which is the only EoS that has been shown to work for asphaltenes in reservoir fluids ranging from condensates to heavy oils.

A “universal workflow” has been developed to measure fluid gradients by DFA and interpret the gradients utilizing both the cubic EoS for gas-liquid equilibria and the FHZ EoS for solid-solution equilibria. Nonequilibrium oil columns that are frequently encountered can be so treated provided the origin of the disequilibrium is identified. A particularly interesting observation in reservoirs is the existence of upstructure bitumen deposition in some reservoirs and tar mat at the oil-water contact in other reservoirs. For many reservoirs, this has been traced to the rate of late gas charge which acts to destabilize asphaltenes. Slow rates of gas charge allow asphaltenes to accumulate at the diffusion front creating density inversions with subsequent convective gravity currents that are generally quite rapid. In contrast fast gas charge gives rise to gas-oil contact displacement that sweeps past original oil leading to upstructure deposition of bitumen. The limiting case of this process is miscible gas injection in EOR.

The resulting understanding of the petroleum system concepts and the thermodynamic and fluid mechanic principles in play in reservoirs has enabled identification of durations of processes that take the reservoir fluids towards thermodynamic equilibrium. These principles are confirmed when projecting the state of the reservoir fluids onto a timeline that incorporates the reservoir age while including subsequent important alterations to the fluid column. Each reservoir condition is associated with important and specific production concerns for optimization of the
field development plan. The concepts presented herein can be used at the earliest stages to guide formation evaluation, even using seismic interpretation as an important component. These concepts do not require a deterministic understanding in order to be useful to reservoir asset teams. Instead, these concepts can be used in a probabilistic sense to narrow the range of key concerns that must be addressed in the evaluation of the reservoir. In this manner, the efficiency of reservoir evaluation can be improved greatly by employing workflows and heuristics that have been presented herein. Reservoir connectivity is first among equals as a major reservoir concern and has been shown repeatedly to be successfully addressed by these new methods; this bodes well for continued success in addressing the unlimited peculiarities reservoirs exhibit.

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