

Understanding the Viscosity of Hydrocarbon-based Liquids

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ABSTRACT

A framework is described for quantitatively understanding and predicting the compositional and temperature dependence of the viscosity of hydrocarbon-based liquids, based on the Modified-Arrhenius equation which captures behavior associated with the approach to the glass transition, even at temperatures well above it. The framework is shown to apply across a broad range of materials, including bitumen, asphalt, lubricants, vegetable oils, and polymer solutions used as viscosity modifiers. When the viscosity–temperature relationship, is parametrized as $\eta(T) = \eta_{\infty} \exp\{D/[(T/T_0) - 1]\}$ with a fixed fragility (D), the two parameters, T_0 and η_{∞} vary systematically with composition and chemical changes such as oxidation. Importantly, this parameterization leads to simple linear blending rules for T_0 and η_{∞} which predict the highly nonlinear, and even non-monotonic viscosity behavior observed in blends.

INTRODUCTION

General prediction of viscosity, especially on blending, has always been difficult. This author's research on viscosity began in the early 1990s with the challenge of developing a fundamental mechanistic understanding, in order to predict and control the viscosity of extremely heavy asphaltene-rich bitumens. Over four decades, that research expanded to include oils of all kinds, from crude oil to vegetable oils. A key enabler was working at the interface of physics, chemistry, and engineering, interacting with technical experts from all parts of our energy and chemicals businesses. But most important has been working in an organization which recognizes the importance of (1) fundamental scientific understanding of our technologies; (2) engagement with the outside scientific community; and (3) the long-term commitment to such research.

As far as the scope of the problem, both from data in the literature and from within our company (ExxonMobil), it was obvious that the problem at hand was not about non-Newtonian shear-dependent rheological behavior, but simply viscosity. Other effects come in when, for example, polymers are added, or when a second phase is present, particularly wax particles which crystallize at lower temperatures forming gel-like structure and a yield-stress. However, that is a different phenomena which can be explained separately (but still requiring knowing the Newtonian viscosity of the continuous liquid phase). Too often when multiple phenomena and mechanisms are present and not separately recognized and modeled, understanding the system can be intractable.

Experimentally we found evidence of viscosity drop associated with increased shear rates, which is often confused with non-Newtonian behavior. Too often, viscosity studies are done isothermally, i.e. characterizing the viscosity of different feeds or blends at a single temperature. However, it is essential to measure and be aware of the temperature dependence, since shear generates heat within the sample, especially in highly viscous liquids. Since rheometers control the temperature at the Couette or plate surfaces where the temperature is usually measured, shear will give rise to a temperature gradient *within* the liquid, where the sample temperature can be significantly above the wall/measured temperature. So, in asphalts, which in the absence of polymer addition are Newtonian, but where the viscosity can drop an order of magnitude over a 10°C rise in temperature, this can be a very large effect.

For oils, in practice, viscosity is all about temperature. This is unlike colloidal systems, where viscosity is essentially athermal, with the temperature-dependence only arising from the continuous liquid phase. Temperature dependence must be front and center in measurement and modeling of oil viscosity. In fact, a 6 order-of-magnitude variation of heavy bitumen viscosity with temperature (over the relevant temperature ranges) is comparable to its variation isothermally by dilution with low-viscosity solvent.¹

More fundamentally, viscosity is about volume or free-volume.^{2, 3, 4} The ease of moving around depends on how much empty space there is. In a very crowded cocktail party, you can't move around and mingle. In a lattice model analogy with a "sliding tile puzzle", if there were no empty space nothing could move. With two empty spaces it would be much easier than with one empty space. When free volume disappears, diffusion stops, resulting in a glass transition.

Pressure couples directly to volume, and very-high (GPa) pressures are needed to study this.⁵ But practically, these pressures are not readily accessible and "free volume" cannot be directly measured. Temperature is easy to measure and control, and is, in practice, the most important lever on viscosity and it links to volume via the thermal expansion coefficient. Therefore, we use the formulation of the free-volume based concepts without parametrizing free-volume explicitly, in the form of the modified-VTF Vogel-Tammann-Fulcher or modified-Arrhenius equation,⁶ (which is mathematically equivalent to the Williams-Landel-Ferry WLF equation).

$$\eta(T) = \eta_{\infty} \exp\{D/[(T/T_0) - 1]\} \quad (1)$$

Single-phase viscosity of oils follows the Modified Arrhenius form (eq 1).¹ There are three parameters: T_0 , η_{∞} and D . Figure 1 illustrates the equation's behavior and what the parameters represent. T_0 , the Kauzmann temperature, is the asymptotic temperature where the viscosity diverges. The glass transition or T_g , is viscometrically defined as where the viscosity reaches 10^{13} Poise, so T_g is connected to T_0 , but a little higher. η_{∞} is the high-temperature limiting viscosity, and D controls how the viscosity varies between those limits.

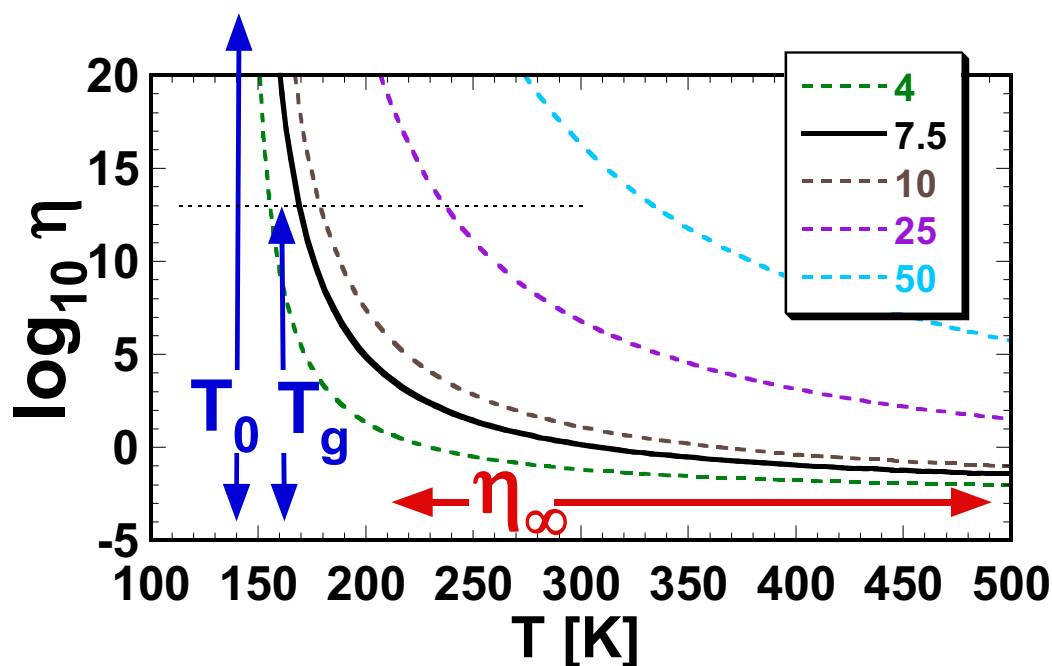


Figure 1: $\eta(T)$, illustrating the asymptotic behavior and meaning of the parameters of the modified-VTF equation, here plotted for $T_0=140$, $\eta_\infty=0.002$, $D=7.5$, and showing T_g . Other values of D are listed in the legend and are shown with thin dashed curves. *Adapted with permission from Ref¹. Copyright 2025. ACS.)*

Measured viscosities as a function of temperature are just part of this curve; and informed by the glass transition even at temperatures far above it. Viscosity data, even with multiple orders-of-magnitude variation, only cover a relatively small fragment of the curve, and it is extremely difficult to uniquely independently determine all three parameters accurately. Therefore, free fits do not give systematic results, as they should for systematic variations in composition. Using a large data set allowing D to float, $D=7.5$ was chosen from its average value, “arbitrarily” as the song says.⁷ And fixing that value did not noticeably degrade the fit quality. Figures 2(a) and 4 show typical data sets fit to the two free parameters, obtaining essentially perfect fits over a wide viscosity range; and most importantly as discussed below, the variation of T_0 and η_∞ are systematic and sensible. When data does not fit this form, it is usually experimental error, shear induced heating, or the appearance of a 2nd phase due to wax crystal formation. The use of this functional form itself is not new. Fixing D , was the critical step.

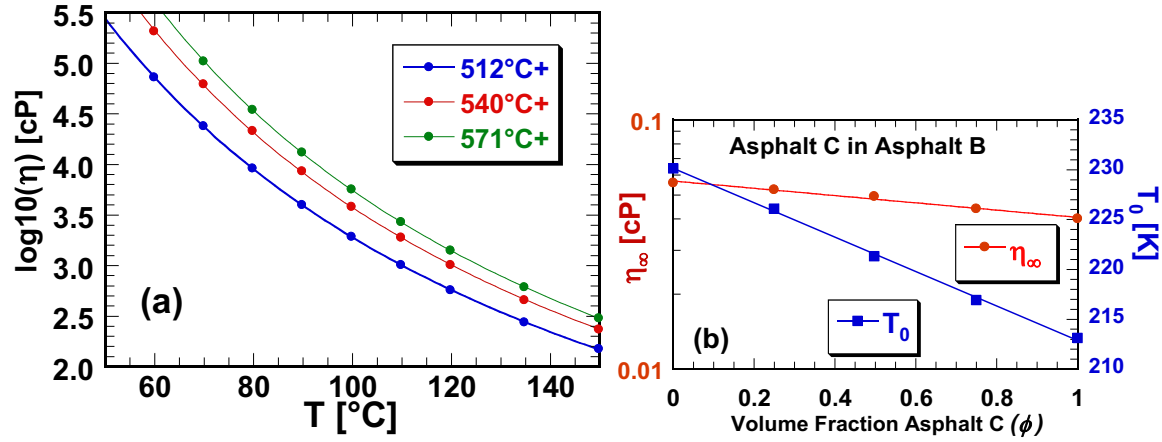


Figure 2: (a) $\eta(T)$ of Asphalt A, a vacuum resid from a typical crude oil, for different distillation cuts. The fit with $D=7.5$ passes through all the points and is shown with solid lines. (b) Typical blending behavior of T_0 and η_∞ derived from $\eta(T)$ of binary blends plotted vs. volume fraction ϕ , shown here for a pair of asphalts.

One doesn't need many data points to get T_0 and η_∞ . As long as one has two trusted viscosities (η_a at T_a , and η_b at T_b) at two temperatures that are not too close together, the parameters can be easily calculated analytically:

$$\begin{aligned} y &\equiv -[D(T_a - T_b)/\ln(\eta_a/\eta_b)] + (T_a + T_b), \\ T_0 &= (y - \sqrt{y^2 - 4T_a T_b})/2 \\ \eta_\infty &= \eta_a \exp\{-D/[(T_a/T_0) - 1]\} \end{aligned} \quad (2)$$

With this parametrization, T_0 and $\ln(\eta_\infty)$ essentially blend linearly with volume fraction, examples of which are shown in Figures 2(b), 3(a) and 4. Generalized, for multiple components:

$$T_0 = \sum_i \phi_i T_{0,i} \quad \ln \eta_\infty = \sum_i \phi_i \ln \eta_{\infty,i} \quad (3)$$

These are not blending rules for viscosity at such-and-such a temperature. These two parameters give the blending behavior at *all* temperatures. And the highly non-linear, and sometimes non-monotonic blending behavior of viscosity at a given temperature is simply explained without resorting to complicated empirical blending indexes or ad-hoc corrections.

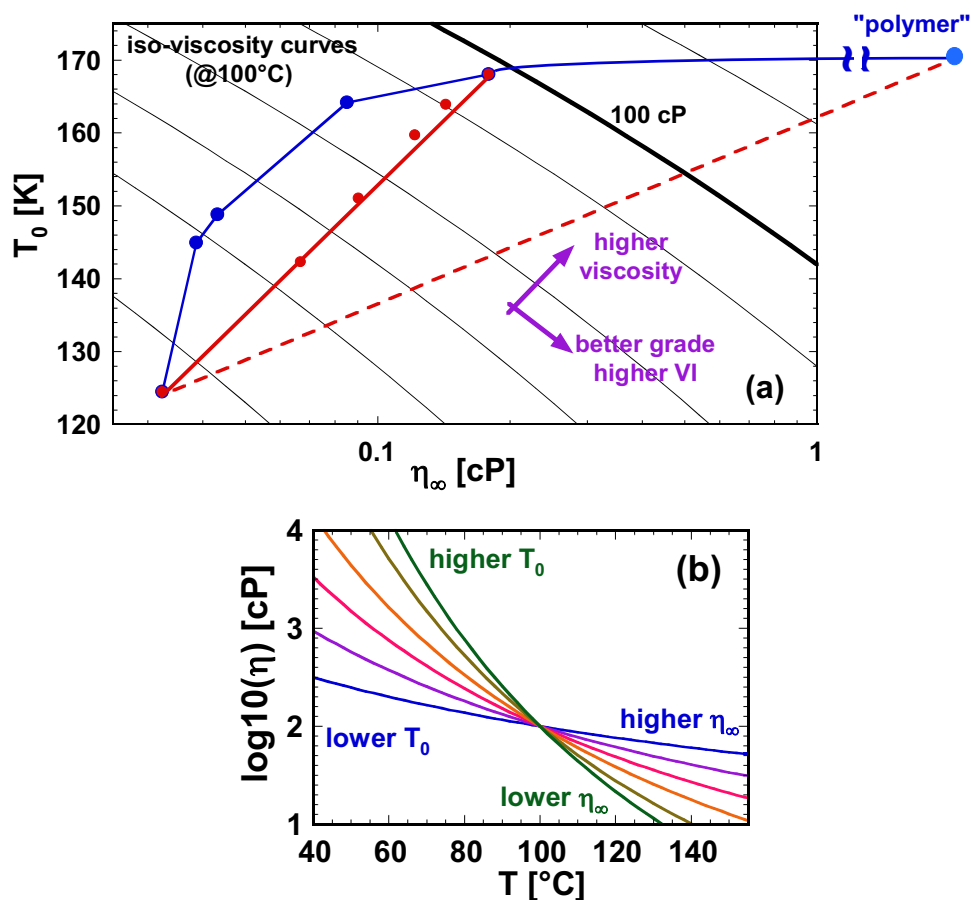


Figure 3: (a) T_0 - η_∞ map where mixtures will lie on lines between their components. The direction of higher viscosity grades and better quality are shown with the purple arrows. Measured values for a series of pure PAOs of increasing MW and viscosity grade are shown with the blue points. Measured blends of the lightest and heaviest are the red points. The blends fall to the lower right of the pure components, meaning smaller sensitivity of viscosity to temperature. The thin diagonal back curves are iso-viscosity curves at 100°C, and run diagonally, with the 100cP curve bolded. The extension to the right representing the much higher MW regime, is schematic and shows the efficient thickening effect of a polymer viscosity modifier. **(b)** $\eta(T)$ for (η_∞, T_0) along the 100cP@100°C iso-viscosity curve.

It's very insightful to look at a T_0 - η_∞ map with a logarithmic η_∞ axis, as shown in Figure 3(a). To understand it, we superimpose iso-viscosity curves for a given temperature (100°C here), with the 100 cP curve bolded. To get a given viscosity at a given temperature, there is a locus of (T_0, η_∞) pairs which will fulfill that, which comprise the iso-viscosity curves. Every oil or mixture has a coordinate on such a plot. $\eta(T)$ for such pairs that give 100cP

@100°C are shown in the Figure 3(b). One can not only calculate, but visualize the blending behavior, since with this parametrization a mixture will lie on a straight line between the components. Regardless of type of product, be it asphalt or lubricants, one can populate the map with different possible components and can therefore get to any target viscosity-temperature behavior by choosing what, and how much, to blend. Following an iso-viscosity curve towards the lower right is lower- T_0 and higher- η_∞ . The purple arrows point to the directions of higher viscosity grades, and towards better quality.

When you look at viscosity this way, many apparently “anomalous” behaviors are simply explained. For example: Some blends exhibit non-monotonic viscosity behavior: At a constant temperature, the viscosity goes down and then up on varying concentration (ϕ). If one component has a higher T_0 (meaning higher viscosity at lower T) and the second has with a higher η_∞ (giving higher viscosity at higher T), then $\eta(T)$ curves will cross at some temperature, as in Figures 4(a,c). Since there are different functional dependences of viscosity on T_0 and η_∞ near that temperature, the viscosity cannot be constant with ϕ , and thus must exhibit extrema. Therefore, the non-monotonic behavior is not an anomaly, but rather an intrinsic consequence of the simple theory.

The behavior of blends of an 8 cSt poly-alpha-olefin (PAO) with both Naphthenic acids and Canola Oil (triglycerides with three ~18-carbon chains, MW~880) are shown in Figure 4. Blends of 8 cSt PAO with Naphthenic Acids (Baker CAS No:1338-24-5) and (Canola oil. $\eta(T)$ for the pure components are shown in Figure 4(a,c) with the higher T_0 component shown in pink and the lower T_0 one in green. The insets show the viscosity as a function of concentration, at the temperature where the $\eta(T)$ curves cross. Figure 4(b,d) shows T_0 and η_∞ derived from $\eta(T)$ of binary blends, plotted vs. volume fraction ϕ . Different functional dependences on T_0 and η_∞ result in extrema and thus non-monotonic behavior on blending, at temperatures near the $\eta(T)$ crossing.

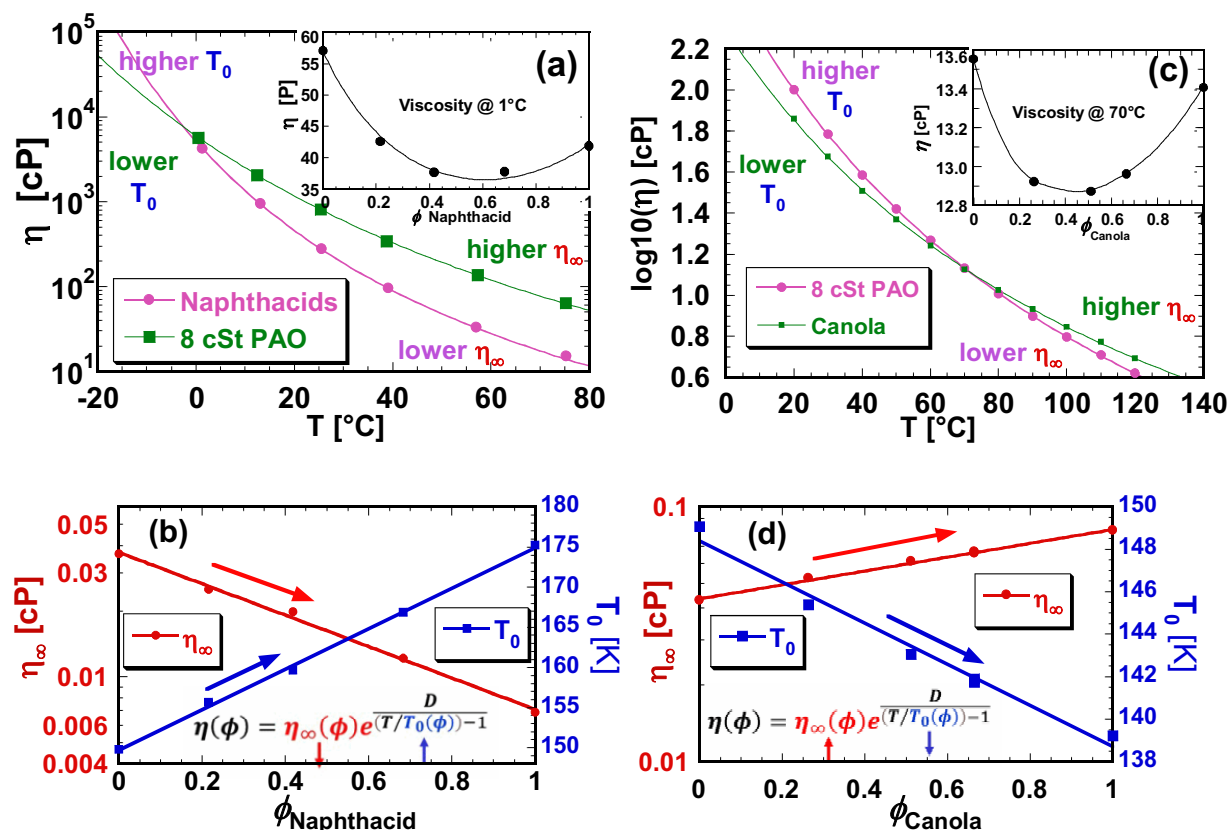


Figure 4. Blends of 8cSt PAO with (a,b) Naphthenic Acids (Baker CAS No:1338-24-5) and (c,d) Canola oil. (a,c) $\eta(T)$ for the pure components with the higher T_0 component shown in pink and the lower T_0 one in green. The insets show the viscosity as a function of concentration, at the temperature where the $\eta(T)$ curves cross. (b,d) T_0 and η_∞ derived from $\eta(T)$ of binary blends plotted vs. volume fraction ϕ . Different functional dependences on T_0 and η_∞ result in extrema on blending when T is near the $\eta(T)$ crossing. (a,b) adapted with permission from Ref¹. Copyright 2025. ACS.)

Parametrized this way, dependences on chemical composition appear systematic.¹ For example, T_0 for the paraffins and other aliphatics is low and initially increases with MW but levels off in the high-MW polymer limit, as we know T_g does for polymers. T_0 is higher with ring-containing species, polar functionalities, and other characteristics associated with higher T_g . η_∞ increases with, and depends mostly just on the MW, continuing to increase strongly with MW. This dominates the high viscosity of polymers, especially polyolefins; This is different than the comparably lower-MW petroleum components where high viscosity is dominated by high T_0 .

Lubricants: A better lubricant's viscosity is less sensitive to temperature. It needs to be pumpable in cold weather so you can start your car on a cold day; but it should not lose its lubrication performance at high temperature. The Viscosity Index or VI, is an industry property specification designed to quantify this.⁸ High VI means “better”, i.e. less sensitive to temperature. VI was historically defined as a piecewise continuous function of the viscosities at 40 and 100°C. It exhibits anomalous blending behavior, which was also simply explained¹ by considering the parameterization of T_0 and η_∞ , rather than kV100 and VI as is common in the industry. So, in the opinion of the author, one should not use VI within R&D or in doing science. Try to understand T_0 and η_∞ ; not VI, because you can't. You can always map back to VI.

It's known that extreme-bimodal blends of synthetics make higher quality lubricants.⁹ Why? For the increasing MW grades, T_0 levels off and η_∞ continues to rise, thus forming a curved path on a T_0 - η_∞ map as in Figure 3. Comparing the pure components to the blends of the extremes for a given viscosity grade, the blends are lower and to the right. That is a weaker dependence on temperature and thus better quality. The extreme bi-modal blends will be to the lower right by simple geometry and have a higher VI than a single component (with equivalent 100°C viscosity). No special explanation needed; it comes right out of the theory.

Polymers: This simple framework is not applicable to entangled high-MW polymer melts where other physical mechanisms leading to complicated rheology occur. However, we can consider dilute or semi-dilute polymer solutions in the Newtonian limit.

As we also needed to do with asphaltenes, we must dispense with thinking in terms of Intrinsic Viscosity $[\eta]$, and picture of polymers in solution following the Einstein relation (shown schematically in Fig. 5(a)) behaving colloidally as hard spheres, which they are not; and as it cannot account for the temperature-dependence associated with the polymer. One should consider $[\eta]$ as the $\phi \rightarrow 0$ limit of the blending of T_0 and η_∞ . Often reported slope changes or breakpoints of the viscosity as a function of concentration at c^* are an artifact of plotting constant-temperature viscosity on a linear scale, assuming the Einstein relation for colloids. Shown in Figure 5 is the rising viscosity as a function of concentration, calculated for solutions of a $T_0=202\text{K}$ material and $T_0=70\text{K}$ liquid. In Figure 5(a) the viscosity is plotted on a linear axis where an apparent break from the linear behavior is often described in the colloidal framework.

However, increasing the scale of the viscosity shows the apparent breakpoint shift to lower concentrations, while on the logarithmic scale (Figure 5(b)) these continue to increase linearly and smoothly; no breakpoint.

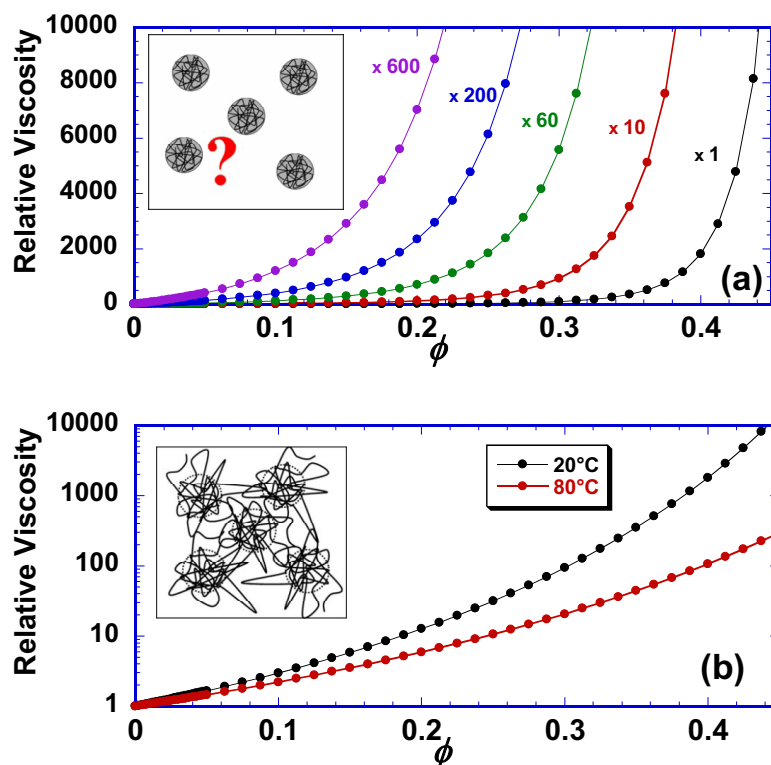


Figure 5: Calculated viscosity upon blending a $T_0=202\text{K}$ material into a $T_0=70\text{K}$ liquid. **(a)** At 20°C, with the curve shown scaled by a series of larger factors, giving an apparent earlier breakpoint to a non-linear rise. Schematic representing soluble polymers as solid colloids. **(b)** Shown on a logarithmic viscosity scale, which shows continuous behavior, for 20°C as well as 80°C. Schematic representing soluble polymers as soluble polymers.

Figure 6 shows η_∞ and T_0 derived from $\eta(T)$ measured on solutions of an ethylene-propylene copolymer ($M_w = 99 \text{ Kg/mol}$, 72% C_2) in Pristane (2,6,10,14-Tetramethylpentadecane, $\text{MW}=268.5$). They exhibit the linear blending behavior that we regularly observe, as described above. Extrapolating T_0 and η_∞ to 100% polymer will give an “effective” T_0 and η_∞ for blending them in solution; but does not simply extrapolate to neat polymers or concentrated solutions where the polymers are entangled.

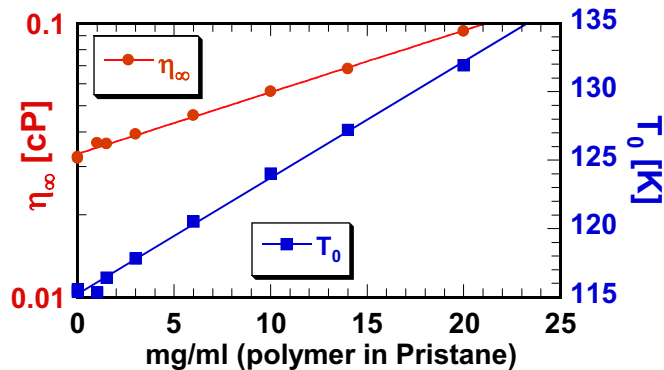


Figure 6. η_{∞} and T_0 of ethylene-propylene copolymer ($M_w = 99000$, 72% C_2) solutions in Pristane as a function of polymer concentration.

This approach easily explains the sometimes-observed negative Intrinsic Viscosities,¹⁰ which are essentially a low- T_g (low- T_0) polymer “plasticizing” the higher T_0 solvent.¹¹

Besides providing a new approach to describing and quantifying polymer solution viscous behavior, soluble polymers are practically important as viscosity modifiers or VI improvers for lubes, asphalt and other oils.^{12, 13} As the MW becomes very large, η_{∞} continues to increase, as shown in Figure 7. Here, the polymer η_{∞} values are recalculated from the polybutadiene data of Colby et al.¹⁴ and the low-MW points are data on n-alkanes from the DIPPR database.¹⁵ For low-MW polymers, the viscosity is known to increase nearly linearly with M_w , while at high-MW when they are entangled, it increases as $\eta \propto M_w^{3.4}$.^{16, 17} The crossover occurs at M_c where entanglements set in.^{18, 19} However, entanglements are suppressed in dilute solutions of polymers, where $M_c(\phi) \sim M_c/\phi$, extending the linear regime.²⁰ So the rise in effective- η_{∞} as a viscosity modifier in dilute solution would more likely be that associated with the extrapolation of the lower MW slope, since the high exponent is due to entanglements which would not be present in dilute solution.

Looking at the schematic “polymer” point in Figure 3(a), which is very high η_{∞} , we see the viscosity grade can be strongly increased while improving quality. Thus, the mechanism responsible for the behavior of viscosity modifiers is the same phenomenon that improved properties through extreme bi-modal blends.

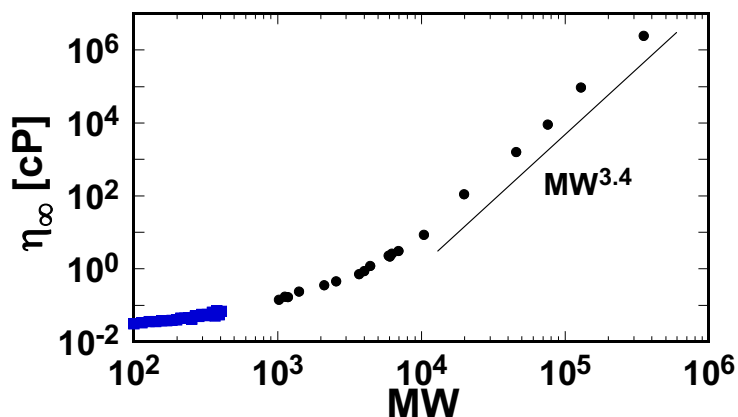


Figure 7. η_{∞} and vs. MW (black) recalculated from the polybutadiene data of Colby et al.¹⁴ and (blue) n-alkane data from DIPPR¹⁵.

While polymer data is not always acquired and/or presented with multiple temperature data points, in some cases it is parameterized using the Modified Arrhenius (VTF) equation (or its equivalent). Even if the raw data is not available, knowing the temperature range over which it was measured (i.e. where that equation represents the data), one can re-parametrize the results with a constant D . When the WLF equation is used, it can be easily mapped to the modified-VTF parametrization. Polymer viscosities are sometimes reported using the Melt Index (MI), which can be related to a nominal low-shear Newtonian melt viscosity $\eta = 80000 \text{ Poise} / \text{MI}$.²¹ Additionally, for high-MW polymers where T_0 has already leveled off, a viscosity measured at a fixed temperature will yield the MW dependence of η_{∞} . Systematically looking at the viscosity of polymers this way would be very insightful.

Asphalt: Asphalt binder contains asphaltenes and is a non-fuel disposition of petroleum vacuum resid. Asphaltenes are molecules in an aromatic-rich, relatively high-MW fraction of petroleum, and are defined by solubility, e.g. toluene-soluble/heptane-insoluble. Their prevalence imparts especially high viscosity to heavy crude oils, bitumen and asphalt. They are usually problematic, but are a beneficial component of asphalt. Asphaltene molecules are usually soluble in crude oil and they are not colloidal dispersions. Understanding asphaltenes as molecules in a highly non-ideal solution, and overcoming the prevalent notion in the literature of “asphaltenes as colloids”

or particles (or “nano-aggregates”), enabled us to make significant strides in understanding, and predictions of both viscosity and phase behavior.^{22, 23, 24}

Asphalt specifications^{25, 26} are essentially designed to ensure that the road should not rut under the weight of heavy trucks on a hot day, and should not be brittle (or glass-like) and crack in cold weather. So “better” asphalt (for a given viscosity or hardness grade) means weaker viscosity sensitivity to temperature, just like for lubricants. Asphalt properties measured through specification test methods are manifestations of actual fundamental physical properties. Most asphalt properties are simply manifestations of viscosity on the $\eta(T)$ curve. For example, the softening point is the temperature at which the sample yields under specific test conditions, which can be associated with reaching a given viscosity (~ 14000 cP).¹ The Penetration is measured as the distance travelled by a needle under a given force, in a given time which is also a measure of viscosity.²⁷ So, if you know T_0 and η_∞ , then you could predict most asphalt properties.

One can put asphalt components on a $T_0 - \eta_\infty$ map like Figure 3(a), where to the upper right is a “harder” or more viscous asphalt, and towards the lower right is a “better” asphalt in terms of sensitivity to temperature. Linear blending on such a map can enable selecting a blend with a chosen property target.²⁸

Physical and chemical changes causing highly non-linear changes to viscosity, behave systematically within this framework and can be represented as paths on the map, shown in Figure 8(a). The compositional path taken on distillation of asphalt to higher cutpoints moves primarily linearly and most importantly to higher T_0 , but also slightly increased η_∞ . We point out that this turns out to be linear over a finite boiling point range but actually depends on the actual (η_∞, T_0) of the components as a function of boiling point.

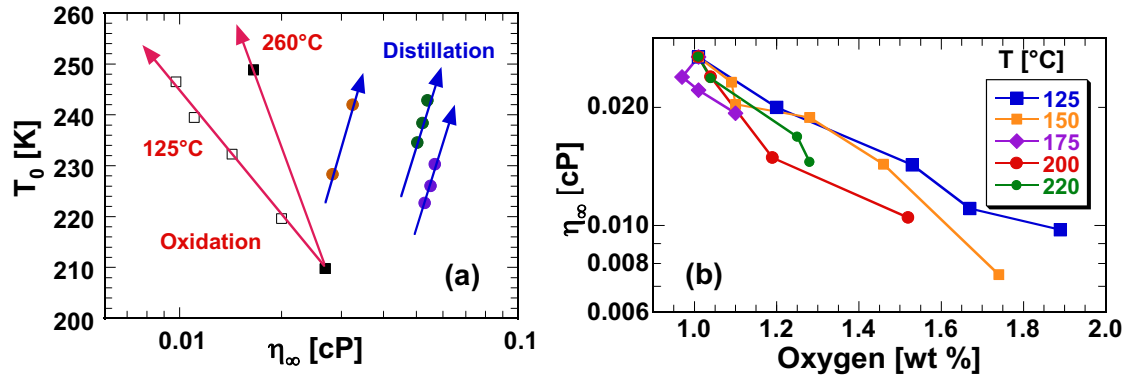


Figure 8: (a) T_0 - η_∞ map showing the paths taken upon distillation to make higher cutpoint vacuum residues from 3 different crude oils (circle, with blue arrows); and the oxidation paths (red arrows) for both higher-temperature (air blowing) and lower-temperature (aging) conditions. While T_0 increases for both, the low-temperature process decreases η_∞ more significantly; thus requiring a higher T_0 to reach the same viscosity of a soft asphalt oxidizing at high- (solid square) and low- (open squares) temperatures, where different chemistries dominate. (b) η_∞ for an asphalt oxidized at various temperatures ranging from 125 to 220°C, each for multiple durations. Further details on this experiment can be found in [1].

Asphalt oxidation occurs in the hot mix plant; at lower temperatures due to aging on the road; as well as during air-blowing, a high-temperature process, sometimes used in asphalt manufacturing to increase its viscosity. While industrially considered totally different things, scientifically we studied them together with a continuum of oxidation temperatures. Viscosity at a reference temperature is a highly non-linear function of oxidation severity. But T_0 exhibits a simple linear increase with oxidation time at temperature, where the rate of increase increases with temperature and can be understood in terms of chemical reaction activation energies.¹ (Figure 8(a)) Along with systematic changes to η_∞ , this can enable prediction of the oxidized or aged property changes from the initial asphalt's properties.

High pressures are relevant to boundary lubrication. While viscosity is a highly non-linear function of pressure, T_0 increases linearly and systematically allowing simple functional forms for the temperature and pressure dependence of viscosity at any temperature. Pressures directly couple to free volume, and T_0 , the largest lever on viscosity, increases roughly linearly with pressure.

CONCLUSIONS

This framework is immensely useful; But everything is not so simple:

There can be non-linearities in the blending of T_0 . Non-linearities are usually negligible, but there are cases where they can be observed with careful measurement. We understand this in terms of Excess Volume of mixing. This means effectively adding “free volume” when two liquids are mixed.

Oxygen content seems to reduce η_∞ . Shown in Figure 8(b) is the measured η_∞ for oxidation of a 390°C⁺ cutpoint asphalt, by bubbling air, with oxidation temperatures ranging from 125 to 220°C for multiple durations ranging from 60 to 3000 minutes. While the reaction rate increases by an order-of-magnitude over this range,¹ the dependence of η_∞ on oxygen content is independent of oxidation temperature, with η_∞ decreasing systematically. We also note that alcohols have lower η_∞ than comparable alkanes. The trend is systematic. But a correlation is not the answer, it poses a question: Why?

As the song⁷ says, “The third parameter is called D . We set (at 7.5) arbitrarily.” There must be compositional dependence to D . What is it? How would one incorporate a variable D in a way that maintains the meaningfulness of T_0 and η_∞ ? And how would that parameter blend?

In summary: Practically, parametrizing viscosity this way simplifies the complex. It’s applicable for oils, well-beyond the energy industry. It is useful. It should be systematically and routinely applied to data, which will be able to teach us a lot and relate chemical composition to viscosity. There is still more to be scientifically understood.

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