

## Viscosity data for kukersite shale gasoline fractions

Zachariah Steven Baird, Alexey Yanchilin, Vahur Oja, Oliver Järvik\*

Department of Energy Technology, Tallinn University of Technology, Ehitajate tee 5, 15006 Tallinn, Estonia

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**Abstract.** *Despite the fact that the shale oil industry in Estonia is a century old, there is still too little data about the thermodynamic and transport properties of the fuel. Information about these properties is important in producing and handling the fuel and in environmental and safety analyses. The current article addresses this problem by presenting experimental viscosity data for the lighter portion of kukersite shale oil, often called shale gasoline. The data is for 23 narrow boiling fractions, and also shows the change in viscosity for portions of the gasoline with different average boiling points. Viscosities were measured over a range of temperatures, and equations were fit to the data to describe the temperature dependence of the viscosity. Correlations are also presented which can be used for predicting the viscosity of kukersite shale gasoline if its density at 20 °C is known.*

**Keywords:** *viscosity, Estonian oil shale, kukersite, experimental data, correlation.*

### 1. Introduction

Viscosity is an important property of a fuel that gives information about its flow behavior. Thus, viscosity data is useful in processing and using liquid fuels. Additionally, viscosity can be used in correlations to calculate other properties of the fuel [1].

Although shale oil has a long history of production and use, data about the thermodynamic and transport properties of shale oils is limited. Shale oil is an alternative fuel that is produced from oil shale via pyrolysis [2, 3]. Oil shale resources are abundant throughout the world [4], and it has been estimated that 4.7 trillion barrels of oil could be produced from the reserves [5]. However, technical and environmental challenges have generally limited the production of shale oil, although active shale industries do exist in several countries, including Brazil, China, and Estonia [5]. In Estonia, shale oil has

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\* Corresponding author: e-mail [oliver.jarvik@taltech.ee](mailto:oliver.jarvik@taltech.ee)

been produced from kukersite oil shale for about a century, but only a small amount of experimental data can be found about its thermodynamic and transport properties, such as viscosity [6]. Most of these data were measured between 1920 and 1950 for shale oils from older types of retorts not used nowadays. Both Kogerman [7] and Kollerov [8] published information about the viscosity of kukersite shale oil, but these were mostly for fractions from higher boiling ranges. Only a few of the samples came from the lighter portion of the shale oil, which is often termed shale gasoline.

To fill this gap, we have measured the viscosities of shale gasoline fractions. Additionally, the shale gasoline was obtained from a plant using the newer solid heat carrier technology, which did not exist at the time of those literature sources mentioned. These measurements were part of a larger project to investigate the thermodynamic and transport properties of kukersite shale oil, with the goal of developing methods for predicting those properties [9]. This experimental data and corresponding predictive correlations provide important resources that can be used in process design and environmental and safety analyses.

## 2. Experimental methods

### 2.1. Sample preparation

The shale gasoline was obtained from Eesti Energia's Narva Oil Plant (Narva, Estonia), which uses Estonian kukersite oil shale. The solid heat carrier retorting method, sometimes called the Galoter process in older literature, is used in this plant [10]. Samples were from the older Enefit 140 plant, also known as UTT 3000.

The wide fractions from the plant were further separated using distillation to get fractions with narrow boiling ranges. One distillation was a simple batch distillation at atmospheric pressure (the Engler distillation [11]) and the other was performed in a rectification column. The properties of both the whole shale gasoline samples and the narrow distilled fractions were given in an earlier article by Järvik et al. [9] or in the Open Science Framework project for this study (<https://osf.io/3q5ur/>). In the current article, we use the same sample identifiers as that earlier article to allow the data to be combined.

The first letter in the identifiers generally designates the fuel sample obtained from the shale oil plant (e.g. G for gasoline fraction). The second letter specifies the type of distillation method used to separate the wide plant sample into narrow fractions (R = rectification, D = batch distillation). The numbers before the dash give the date the distillation was performed, and the number following the dash gives the fraction number.

## 2.2. Viscosity measurements

Viscosity was measured using a capillary viscometer. The capillary viscometer was submerged in a glycerine bath in order to regulate the temperature. 23 shale gasoline samples were measured, and all were measured at multiple temperatures. The expanded uncertainty of the capillary viscometer was  $\pm 0.3\%$ . The measured viscosities spanned the temperature range of  $-10$  to  $160$  °C. The specific range measured for each sample varied.

## 2.3. Other characteristic data

The dynamic viscosity was also calculated using measured density data. Densities were measured using a DMA 5000 M density meter (Anton Paar GmbH, Graz, Austria). The estimated standard uncertainty of the densities measured using this device was  $0.00015$  g/cm<sup>3</sup>. The density data was presented in the earlier article by Järvi et al. [9].

Average boiling points were calculated for the rectification samples as the average of the distillation temperatures of each fraction. Average molecular weights were calculated using cryoscopy with benzene as the solvent [12].

## 3. Results and discussion

### 3.1. Experimental viscosity data

The experimental data on the viscosity of kukersite shale gasoline fractions is given in Table 1 and Table 2 and is shown visually in the Figure. The data is also available on Open Science Framework (<https://osf.io/yzcf8/>). It is worth briefly mentioning that rectification uses a distillation column, and therefore, produces fractions with more symmetric boiling point distributions. The Engler (batch) distillation does not provide as clean of a separation and the boiling point distributions can be somewhat skewed.

**Table 1. Experimental viscosity data for shale gasoline fractions separated using rectification**

GR060613-1			GR060613-2			GR060613-3		
Temp., K	Visc., cSt	Visc., mPa·s	Temp., K	Visc., cSt	Visc., mPa·s	Temp., K	Visc., cSt	Visc., mPa·s
266.2	0.4815	0.3461	263.2	0.6012	0.4427	263.2	0.7070	0.5462
269.1	0.4685	0.3354	269.1	0.5647	0.4125	269.1	0.6602	0.5062
278.9	0.4299	0.3034	274.0	0.5371	0.3898	274.0	0.6252	0.4764
288.8	0.3970	0.2762	283.9	0.4899	0.3508	283.9	0.5654	0.4254
293.7	0.3821	0.2639	293.6	0.4506	0.3183	293.6	0.5165	0.3837
			303.2	0.4174	0.2910	303.2	0.4761	0.3492
			313.1	0.3879	0.2666	313.1	0.4396	0.3182
						323.1	0.4082	0.2915

Table 1 (continued)

GR060613-4			GR060613-5			GR060613-6		
Temp., K	Visc., cSt	Visc., mPa·s	Temp., K	Visc., cSt	Visc., mPa·s	Temp., K	Visc., cSt	Visc., mPa·s
266.2	0.7795	0.6155	274.0	0.7326	0.5626	274.0	0.8018	0.6356
269.1	0.7524	0.5921	293.6	0.5948	0.4459	298.3	0.6196	0.4775
278.9	0.6698	0.5207	313.1	0.5003	0.3661	323.2	0.4944	0.3698
288.8	0.6034	0.4634	333.0	0.4295	0.3064	347.9	0.4121	0.2989
293.7	0.5753	0.4391	352.9	0.3751	0.2607	362.9	0.3738	0.2660
303.2	0.5271	0.3975						
323.1	0.4470	0.3285						
343.0	0.3868	0.2769						
GR060613-7			GR060613-8			GR060613-9		
Temp., K	Visc., cSt	Visc., mPa·s	Temp., K	Visc., cSt	Visc., mPa·s	Temp., K	Visc., cSt	Visc., mPa·s
298.3	0.6520	0.5126	298.3	0.6750	0.5230	298.3	0.7174	0.5507
313.2	0.5657	0.4372	313.2	0.5850	0.4455	313.2	0.6193	0.4674
333.1	0.4797	0.3622	333.1	0.4941	0.3676	333.1	0.5214	0.3845
352.9	0.4165	0.3069	352.9	0.4265	0.3098	353.0	0.4491	0.3234
372.8	0.3669	0.2638	372.8	0.3742	0.2652	372.9	0.3945	0.2773
			382.8	0.3538	0.2477	382.8	0.3721	0.2583
GR060613-10			GR060613-11			GR060613-12		
Temp., K	Visc., cSt	Visc., mPa·s	Temp., K	Visc., cSt	Visc., mPa·s	Temp., K	Visc., cSt	Visc., mPa·s
298.3	0.7637	0.6022	298.3	0.8091	0.6498	298.3	0.8391	0.6709
313.2	0.6553	0.5084	313.2	0.6926	0.5475	313.2	0.7141	0.5618
343.0	0.5073	0.3804	343.0	0.5310	0.4061	333.1	0.5919	0.4557
372.8	0.4116	0.2981	372.8	0.4286	0.3168	372.8	0.4377	0.3222
392.7	0.3662	0.2589	392.7	0.3797	0.2742	392.7	0.3880	0.2790
			402.7	0.3610	0.2576	407.6	0.3567	0.2520
GR060613-13			GR060613-14			GR060613-15		
Temp., K	Visc., cSt	Visc., mPa·s	Temp., K	Visc., cSt	Visc., mPa·s	Temp., K	Visc., cSt	Visc., mPa·s
298.3	0.8938	0.7105	298.3	0.9714	0.7877	298.3	1.037	0.8512
313.2	0.7562	0.5917	313.2	0.8140	0.6499	313.2	0.8645	0.6987
333.1	0.6234	0.4773	333.1	0.6646	0.5196	343.0	0.6385	0.5004
372.8	0.4588	0.3360	353.0	0.5596	0.4283	372.8	0.5021	0.3811
392.7	0.4014	0.2873	372.9	0.4813	0.3603	392.7	0.4391	0.3260
407.6	0.3693	0.2597				422.5	0.3694	0.2652

**Table 1 (continued)**

<b>GR060613-16</b>		
Temp., <i>K</i>	Visc., <i>cSt</i>	Visc., <i>mPa·s</i>
298.3	1.133	0.9283
313.2	0.9432	0.7615
343.0	0.6845	0.5360
372.8	0.5297	0.4019
392.7	0.4600	0.3416
422.5	0.3748	0.2692
432.5	0.3701	0.2628

**Table 2. Experimental viscosity data for shale gasoline fractions separated using the Engler (simple) distillation**

<b>GD0513-2</b>			<b>GD0513-3</b>			<b>GD0513-4</b>		
Temp., <i>K</i>	Visc., <i>cSt</i>	Visc., <i>mPa·s</i>	Temp., <i>K</i>	Visc., <i>cSt</i>	Visc., <i>mPa·s</i>	Temp., <i>K</i>	Visc., <i>cSt</i>	Visc., <i>mPa·s</i>
274.0	0.6423	0.4879	274.0	0.7309	0.5678	303.2	0.5960	0.4543
293.6	0.5281	0.3914	298.3	0.5658	0.4268	323.1	0.5004	0.3724
313.1	0.4544	0.3283	323.2	0.4596	0.3361	343.0	0.4305	0.3129
333.0	0.3983	0.2803	347.9	0.3866	0.2738	362.9	0.3779	0.2679
352.9	0.3693	0.2530	362.9	0.3595	0.2497	382.7	0.3491	0.2409
<b>GD0513-5</b>			<b>GD0513-6</b>			<b>GD0513-7</b>		
Temp., <i>K</i>	Visc., <i>cSt</i>	Visc., <i>mPa·s</i>	Temp., <i>K</i>	Visc., <i>cSt</i>	Visc., <i>mPa·s</i>	Temp., <i>K</i>	Visc., <i>cSt</i>	Visc., <i>mPa·s</i>
298.3	0.6291	0.4905	298.3	0.6963	0.5526	298.2	0.9454	0.7636
313.1	0.5490	0.4208	313.1	0.6023	0.4704	313.2	0.7945	0.6319
338.1	0.4500	0.3350	338.1	0.4876	0.3704	343.0	0.5955	0.4587
362.9	0.3808	0.2752	362.9	0.4087	0.3017	372.8	0.4729	0.3525
382.8	0.3477	0.2451	382.8	0.3613	0.2605	412.5	0.3715	0.2646
<b>GD0513-8</b>								
Temp., <i>K</i>	Visc., <i>cSt</i>	Visc., <i>mPa·s</i>						
298.2	1.103	0.9064						
313.2	0.9128	0.7389						
343.0	0.6690	0.5252						
372.8	0.5230	0.3977						
412.5	0.4046	0.2944						

### 3.2. Modeling the temperature dependence of the viscosity

Two different equation forms were used for describing the temperature dependence of the viscosity for the shale gasoline samples. One is a double logarithm form given by Seeton [13], and the other is an exponential equation called the Vogel–Fulcher–Tammann (VFT) equation [14]. The equation given by Seeton is an improved version of the more well-known Wright formula [15] and is shown here as Equation (1):

$$\ln \left( \ln \left( \nu + 0.7 + e^{-\nu} K_0(\nu + 1.244067) \right) \right) = A - B \ln(T), \quad (1)$$

where  $\nu$  is the kinematic viscosity in centistokes ( $1 \text{ cSt} = 10^{-6} \text{ m}^2 \text{ s}^{-1}$ ) at temperature  $T$  (in kelvin),  $K_0$  is the zero-order modified Bessel function of the second kind and  $A$  and  $B$  are constants. Note that  $\nu + 1.244067$  is the input to the Bessel function. This equation fits the experimental data with an average relative deviation of 0.99%.

The VFT equation is shown as Equation (2):

$$\mu = A e^{\left(\frac{B}{T-C}\right)}, \quad (2)$$

where  $\mu$  is the dynamic viscosity in mPa·s at temperature  $T$  (in kelvin) and  $A$ ,  $B$  and  $C$  are constants. Although all three parameters are usually fit to experimental data, we found that for shale gasoline fractions the constant  $C$  could be fixed without any significant change in the accuracy of the fit. So,  $C$  was set to be 32 and only  $A$  and  $B$  were fit to the experimental data. This was important because with the simpler equation it was easier to predict the coefficients  $A$  and  $B$ . The VFT equation, with  $C$  fixed at 32, had an average relative deviation of 0.26% when compared to the experimental data. The constants for both of these equations are given in Table 3.

Using the VFT equation, the viscosity curves over the measured temperature ranges were plotted, and this plot is shown in the Figure.

Table 3. Coefficients for the temperature dependent equations fit to the experimental viscosity data

Identifier	Density at 20 °C, g/cm <sup>3</sup>	Average boiling point, °C	Molecular weight, g/mol	Seeton Equation			VFT Equation		
				A	B		A	B	C
GD0513-2	0.741504	–	90	13.478900	2.596276		0.032416	653.374895	32
GD0513-3	0.759058	–	97	15.634884	2.953608		0.026139	744.207382	32
GD0513-4	0.771538	–	102	14.625764	2.764043		0.026546	768.919026	32
GD0513-5	0.784142	–	100	15.088367	2.841698		0.026750	774.203153	32
GD0513-6	0.798023	–	117	16.527204	3.073520		0.024500	830.344400	32
GD0513-7	0.812001	–	123	17.869295	3.254494		0.022420	938.978711	32
GD0513-8	0.825929	–	136	18.844026	3.399160		0.021421	996.071984	32
GR060613-1	0.691195	44.1	69	14.344875	2.819254		0.026210	604.536526	32
GR060613-2	0.706998	65.9	87	15.416471	2.972055		0.025564	659.483297	32
GR060613-3	0.743323	79.9	98	16.274441	3.094125		0.026006	703.968716	32
GR060613-4	0.763755	91.4	105	16.900358	3.181705		0.024452	755.603631	32
GR060613-5	0.750188	101.3	96	16.246343	3.060557		0.024866	755.191952	32
GR060613-6	0.775330	110.5	98	16.697629	3.123560		0.024896	785.093876	32
GR060613-7	0.790917	118.1	101	16.163661	3.022393		0.024730	807.656147	32
GR060613-8	0.779295	123.5	105	16.309729	3.041499		0.023403	828.163690	32
GR060613-9	0.772081	129.4	111	16.455452	3.055572		0.023811	836.901330	32
GR060613-10	0.793081	136.8	107	16.744016	3.094445		0.023999	858.567063	32
GR060613-11	0.807635	143.2	112	16.862700	3.104976		0.024104	877.816783	32
GR060613-12	0.803827	147.6	117	17.094974	3.139551		0.023330	894.549440	32
GR060613-13	0.799272	153.0	118	17.581844	3.213391		0.022502	919.493652	32
GR060613-14	0.815087	161.1	125	19.000799	3.446512		0.022156	950.381032	32
GR060613-15	0.824830	167.8	123	18.357255	3.324449		0.021823	974.986863	32
GR060613-16	0.823615	174.8	131	18.838495	3.394101		0.020167	1020.133560	32

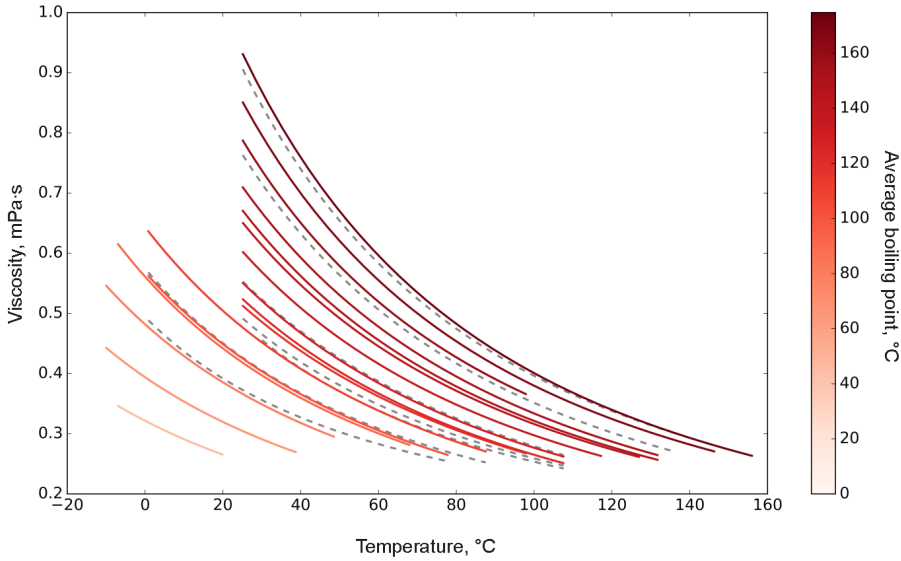


Figure. Viscosities of the shale gasoline fractions. Curves were plotted using the VFT equation and the constants found from fitting the experimental data. The dashed lines are for the samples from the Engler distillation for which the average boiling point was not measured.

### 3.3. Predicting the viscosity of kukersite shale gasoline

The coefficients  $A$  and  $B$  for the VFT equation can be predicted from the average properties of a shale gasoline sample. We created correlations for predicting  $A$  and  $B$  from the sample density at 20 °C. These correlations are given as Equations (3) and (4):

$$A = -0.217907\rho_{20}^{18.5098} + 0.0272718, \quad (3)$$

$$B = 2836.00\rho_{20}^{9.42979} + 531.662, \quad (4)$$

where  $\rho_{20}$  is the density ( $\text{g}/\text{cm}^3$ ) at 20 °C.

When using the coefficients predicted using Equations (3) and (4), the average relative deviation of the predicted viscosities is 4.5%. This is a good result when considering the fact that viscosity is one of the most difficult fuel properties to predict [1]. One likely reason these correlations give good results is that the range of samples and temperatures covered by the data is relatively small. Another is because it was possible to keep the  $C$  coefficient



constant, thereby reducing the complexity of the VFT equation. When the  $C$  coefficient was not fixed, the coefficients did not follow much of a trend versus the density or molecular weight of the samples, which made predicting the coefficients difficult.

These correlations are valid for kukersite shale gasoline samples with densities between 0.691 and 0.826 g/cm<sup>3</sup> at 20 °C, which is the range covered by the samples used. This roughly corresponds to samples with average normal boiling points between about 40 and 175 °C, which is most of the range covered by kukersite shale gasoline. For samples with higher normal boiling points, which are usually then part of the fuel oil, these equations are no longer appropriate. However, an even simpler reason these correlations do not work for heavier fractions is that the  $C$  value for these fractions needs to be higher (approximately 200). A lower  $C$  value does not allow a curve that approximates the temperature dependence of the viscosity for heavier shale oil fractions.

For comparison, we also predicted viscosities using the equation developed by Singh (Equation (8.19) in reference [1]). This equation is for predicting the viscosities of petroleum liquids at different temperatures using only the viscosity of the fuel at 37.8 °C and is shown as Equation (5):

$$\log_{10}(v_{37.8}) = A \left( \frac{311}{T} \right)^B - 0.8696,$$

$$A = \log_{10}(v_{37.8}) + 0.8696, \quad (5)$$

$$\ln(\ln(v + 0.7 + e^{-v}K_0(v + 1.244067))) = A - B \ln(T),$$

where  $v_{37.8}$  is the kinematic viscosity at 37.8 °C, cSt, and  $T$  is the temperature in kelvin. Using this equation the average relative deviation for the shale gasoline fractions was 22%, which is several times greater than the average error of 6% mentioned in [1]. The equation simply did not predict the correct temperature dependence of the viscosity, as could be seen from the fact that the residuals followed a clear trend when plotted versus temperature. This may be due to the difference in composition between shale gasoline and petroleum. Kukersite shale oil, including the lighter gasoline fraction, is more aromatic than most petroleum fractions with comparable distillation ranges [16].

#### 4. Conclusions

This experimental viscosity data for kukersite shale gasoline provides information not currently available in the literature. Additionally, the data was measured for narrow boiling fractions of the shale gasoline and shows the progressive change in viscosity for the different portions of the shale

gasoline. Although the well-known correlation used to predict the temperature dependence of petroleum liquids performed poorly for the shale gasoline fractions, the viscosity for shale gasoline samples can be accurately calculated from their density using correlations developed from the experimental data and presented in the article.

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