resources

MDPI

Article

Different Nonlinear Regression Techniques and Sensitivity Analysis as Tools to Optimize Oil Viscosity Modeling

Dicho Stratiev [1,*], Svetoslav Nenov [2], Dimitar Nedanovski [3], Ivelina Shishkova [2], Rosen Dinkov [2], Danail D. Stratiev [4], Denis D. Stratiev [5], Sotir Sotirov [5], Evdokia Sotirova [5], Vassia Atanassova [5], Krassimir Atanassov [4,5], Dobromir Yordanov [5], Nora A. Angelova [2,3], Simeon Ribagin [5] and Liliana Todorova-Yankova [2]

1 LUKOIL Neftohim Burgas, 8104 Burgas, Bulgaria
2 Department of Mathematics, University of Chemical Technology and Metallurgy, Kliment Ohridski 8, 1756 Sofia, Bulgaria; nenov@uctm.edu (S.N.); Shishkova.Ivelina.K@neftochim.bg (I.S.); Dinkov.Rosen.K@neftochim.bg (R.D.); nora.angelova@fmi.uni-sofia.bg (N.A.A.); litoian@abv.bg (L.T.-Y.)
3 Faculty of Mathematics and Informatics, St. Kliment Ohridski University, 15 Tsar Osvoboditel Blvd, 1504 Sofia, Bulgaria; dnedanovski@fmi.uni-sofia.bg
4 Institute of Biophysics and Biomedical Engineering, Bulgarian Academy of Sciences, Academic Georgi Bonchev 105, 1113 Sofia, Bulgaria; danail.stratiev@gmail.com (D.D.S.); k.t.atanassov@gmail.com (K.A.)
5 Intelligent Systems Laboratory, Department Industrial Technologies and Management, University Prof. Dr. Assen Zlatarov, Professor Yakimov 1, 8010 Burgas, Bulgaria; aitos_dsdd@abv.bg (D.D.S.); ssotirov@btu.bg (S.S.); esotirova@btu.bg (E.S.); vassia.atanassova@gmail.com (V.A.); dobromirj@abv.bg (D.Y.); sim_ribagin@mail.bg (S.R.)
* Correspondence: stratiev.dicho@neftochim.bg

check for updates

Citation: Stratiev, D.; Nenov, S.; Nedanovski, D.; Shishkova, I.; Dinkov, R.; Stratiev, D.D.; Stratiev, D.D.; Sotirov, S.; Sotirova, E.; Atanassova, V.; et al. Different Nonlinear Regression Techniques and Sensitivity Analysis as Tools to Optimize Oil Viscosity Modeling. *Resources* **2021**, *10*, 99. https://doi.org/10.3390/resources10100099

Academic Editors: Ben McLellan and Elena Magaril

Received: 24 August 2021
Accepted: 24 September 2021
Published: 29 September 2021

Abstract: Four nonlinear regression techniques were explored to model gas oil viscosity on the base of Walther's empirical equation. With the initial database of 41 primary and secondary vacuum gas oils, four models were developed with a comparable accuracy of viscosity calculation. The Akaike information criterion and Bayesian information criterion selected the least square relative errors (LSRE) model as the best one. The sensitivity analysis with respect to the given data also revealed that the LSRE model is the most stable one with the lowest values of standard deviations of derivatives. Verification of the gas oil viscosity prediction ability was carried out with another set of 43 gas oils showing remarkably better accuracy with the LSRE model. The LSRE was also found to predict better viscosity for the 43 test gas oils relative to the Aboul Seoud and Moharam model and the Kotzakoulakis and George.

Keywords: vacuum gas oil; gas oil; viscosity; empirical modeling; sensitivity analysis; Akaike information criterion; Bayesian information criterion; nonlinear regression

1. Introduction

The modeling of characteristics of petroleum and its derivatives has been a subject of numerous studies [1,2]. Different regression techniques [3–14] and artificial intelligence [15,16] (machine learning, neural network) approaches have been applied to model petroleum characteristics. Nonlinear regression has been the most used approach for model parameter estimation [17]. Typically, it minimizes an objective function based on the sum of squares of errors between experimental and calculated values [17]. Usually, the models have various parameters to be determined, and sometimes multiple solutions of the objective function can be obtained. The optimal solution depends mostly on the initial guess of parameters [17]. The appropriate parameter estimation has been reported to assure by application of sensitivity analysis on the calculated parameter values [17]. The sensitivity analysis (SA) is the study of how the variation in the output of a model (numerical or otherwise) can be apportioned, qualitatively or quantitatively, to different sources of variation, and how the given model depends on the information fed into it [18].

Good modeling practice requires that the modelers provide an evaluation of the confidence in the model, possibly assessing the uncertainties associated with the modeling process and with the outcome of the model itself [18]. Originally, SA was created to deal simply with uncertainties in the input variables and model parameters. Over the course of time, the ideas have been extended to incorporate model conceptual uncertainty, that is, uncertainty in model structures, assumptions, and specifications [18]. In our recent research [14], we developed an empirical model to predict the viscosity of secondary vacuum gas oils (VGOs) that outperformed the existent empirical models published in the literature. This model was developed based on data for 24 VGOs, extending the model of Aboul-Seoud and Moharam [1] by separating the influence of the specific gravity and the average boiling point on the VGO viscosity, adopting the idea of Kotzakoulakis and George [7]. The model was validated with data for 10 additional VGOs not included in the initial database of 24 VGOs showing a better prediction ability than the model of Aboul-Seoud and Moharam [14]. In that study [14], we applied nonlinear regression using the classical approach for estimation of model parameters by minimization of the sum of squares of errors between experimental and calculated values. The viscosity measurement, however, is associated with a relatively high error (about 5% repeatability, and about 15% reproducibility) [19]. The error in viscosity measurement in our recent study [14] was found to linearly increase with the temperature of the measurement decreasing (between 5.5 and 57.8% for the temperature range 60–100 °C, being the lowest at the highest temperature).

The model parameters can be estimated not only by minimization of the sum of squares of errors between experimental and calculated values but also by minimization of the sum of absolute errors, and by minimization of the sum of relative errors [20]. Which of these nonlinear regression methods gives the best prediction is a question that needs to be investigated. For that reason, we employed data of 41 VGOs from primary and secondary origin to examine the application of four nonlinear regression methods: classical least square method, minimization of the sum of absolute errors, minimization of the sum of the squares of relative errors, and the minimization of the sum of the absolute relative errors for modeling of VGO viscosity prediction with the aim to answer the question which nonlinear regression method provides the most appropriate prediction of viscosity of VGO and other oils.

Hernández et al. [3], Hosseinifar, and Jamshidi [4], Samano et al. [17], and Alcazar, and Ancheyta [21], after the application of nonlinear regression, employed sensitivity analysis to find the most appropriate values of the model parameters. This approach was also adopted in this work and extended not only to the model parameters but also to the given data. In the works mentioned above [3,4,17,21] no sensitivity analysis with respect to the given data has been carried out.

The aim of this research is to evaluate which nonlinear regression technique is best suited to model oil viscosity and how the application of sensitivity analysis with respect to obtained model parameters and with respect to given data can assist in the selection of the most appropriate model.

2. Materials and Methods

2.1. Experimental Materials and Methods

Kinematic viscosity at 80 °C, specific gravity, average boiling point, refractive index, molecular weight, and aromatic ring index of the 43 VGOs from primary and secondary origin were used to develop the empirical model for prediction of viscosity applying the four nonlinear regression methods are presented in Table 1. Kinematic viscosity of VGOs was estimated on the basis of the Engler specific viscosity measured in accordance with ASTM D1665 at 80 °C using Equation (1) [22]:

$$\text{Kin. vis.} = 7.41 \times \text{Engler specific viscosity}, \quad (1)$$

where

Kin. vis. = kinematic viscosity, mm^2/s

Engler specific viscosity = Engler specific viscosity, °E

Table 1. Properties of primary and secondary VGOs used to develop the empirical model for the prediction of viscosity applying the four nonlinear regression methods.

Nr	Sample	SG	$T_{10\%}$	$T_{50\%}$	$T_{90\%}$	$T_{95\%}$	ABP, °C	Kin. vis. at 80 °C, mm^2/s	RI at 20 °C	Kw	MW, g/mol	ARI
1	HAGO-1	0.9512	343	397	455	476	398	7.3	1.5385	11.20	342	2.2
2	LVGO-1	0.9715	343	414	493	517	417	12.1	1.5509	11.07	364	2.5
3	HVGO-1	0.9858	426	491	548	562	488	49.9	1.5524	11.27	461	3
4	HAGO-2	0.959	335	395	458	480	396	13.6	1.5442	11.09	339	2.3
5	LVGO-2	0.9856	330	410	488	508	409	15.2	1.5612	10.86	354	2.7
6	HVGO-2	1.0084	430	489	540	554	486	62.1	1.5685	11.00	458	3.4
7	HAGO-3	0.9514	323	377	439	461	380	12.9	1.5409	11.09	322	2.1
8	LVGO-3	0.9768	324	395	482	508	400	16.7	1.5567	10.91	344	2.5
9	HVGO-3	0.997	405	470	534	551	470	34.8	1.5626	11.05	434	3.1
10	FCC SLO-1	0.9871	232	282	412	455	309	3.6	1.5763	10.29	254	2.4
11	FCC SLO-2	1.0549	292	372	475	518	380	9.9	1.614	10.01	319	3.3
12	FCC SLO-3	1.0573	329	392	471	493	397	16.2	1.6135	10.07	337	3.5
13	FCC SLO-4	1.0671	337	401	476	498	405	21.3	1.6194	10.02	346	3.6
14	FCC SLO-5	1.0624	324	391	471	494	395	17.4	1.6172	10.01	335	3.5
15	FCC SLO-6	1.0953	331	400	491	525	407	33.8	1.6392	9.77	346	3.9
16	FCC SLO-7	1.0788	326	397	493	531	405	24.2	1.628	9.91	345	3.7
17	FCC SLO-8	1.063	317	389	484	520	397	18.5	1.6178	10.01	337	3.5
18	FCC SLO-9	1.0835	327	401	480	501	403	28.5	1.6309	9.85	342	3.8
19	FCC SLO-10	1.177	371	435	562	634	456	312.8	1.6927	9.30	395	5.1
20	FCC SLO-11	1.1011	332	394	482	530	403	21.2	1.644	9.70	340	3.9
21	VGO blend	0.9165	376	446	525	544	449	14.2	1.5088	11.91	404	1.7
22	HAGO-4	0.905	357	425	489	505	424	8	1.5029	11.92	371	1.4
23	LVGO-4	0.912	322	417	528	550	422	8.6	1.5088	11.82	369	1.6
24	HVGO-4	0.922	411	486	552	568	483	27.2	1.5082	12.02	453	1.8
25	HAGO-5	0.9710	338	395	459	480	397	13.0	1.5532	10.96	341	2.5
26	LVGO-5	0.9860	320	391	470	495	394	13.0	1.5642	10.78	337	2.6
27	HVGO-5	1.0150	419	477	531	545	476	57.5	1.5751	10.88	442	3.4
28	FCC SLO-12	1.0970	333	395	487	545	405	22.2	1.6417	9.74	343	3.9
29	VBGO-1	0.9399	376	445	495	505	439	14.7	1.5259	11.56	391	2.1
30	VBGO-2	0.9449	373	433	486	497	431	13.5	1.5307	11.45	381	2.1
31	FCC SLO-13	1.0529	278	366	459	483	368	14.5	1.6139	9.96	306	3.2
32	FCC SLO-14	1.0765	321	386	469	493	392	16.2	1.6283	9.86	330	3.6
33	HTVGO-1	0.8939	364	433	506	521	434	10.41	1.4949	12.13	383	1.3
34	HTVGO-2	0.8901	360	429	504	520	431	9.57	1.4927	12.16	378	1.2
35	BG LIGHT	0.8650	306	376	464	514	382	3.7	1.4786	12.21	319	0.8
36	PEMBINA	0.8940	340	428	522	629	430	7.8	1.4936	12.10	378	1.2
37	EKOFISK	0.9030	342	444	535	577	440	7.8	1.5013	12.04	391	1.4
38	BRENT	0.8940	322	406	502	555	410	8.4	1.4990	11.98	353	1.3
39	BOW RIVER	0.9320	342	421	504	570	422	9.5	1.5171	11.56	370	1.8
40	COKER	1.009	333	429	514	560	425	20.7	1.5761	10.70	374	3.1
41	BU ATTIFEL	0.8380	385	445	512	550	447	8.3	1.4541	13.01	393	0.0

Note: Properties of VGOs under numbers 35–40 were taken from Fisher [23].

The specific gravity of VGOs was measured in accordance with ASTM D 4052 method. The distillation characteristics were measured by high-temperature simulation distillation (HTSD) according to the ASTM D7169 method. The average boiling point was estimated by Equation (2):

$$ABP = \frac{T_{10\%} + T_{30\%} + T_{50\%} + T_{70\%} + T_{90\%}}{5}. \quad (2)$$

2.2. Theory/Calculation

2.2.1. Models

Walther's equation [24] was used as a basis for the empirical modeling of the viscosity of oils [7,10]. Mehrotra [10] proposed a correlation that has the form:

$$ln(ln(\nu + 0.8)) = \alpha_1 + \alpha_2 ln(T), \tag{3}$$

with

$$\alpha_1 = 0.148(T_b)^{0.5} + 5.489 \tag{4}$$

and

$$\alpha_2 = -3.7. \tag{5}$$

Aboul-Seoud and Moharam [1] modified Equations (3)–(5) by including in it oil specific gravity and the empirical model then took the form:

$$ln(ln(\nu + 0.8)) = \alpha_1 + \alpha_2 ln(T), \tag{6}$$

where

$$\alpha_1 = 4.3414(T_b\gamma)^{0.2} + 6.6913 \text{ and } a_2 = -3.7. \tag{7}$$

We started our model development from a form analogous to the modified Walther's equation as shown in Equations (6) and (7), having the following appearance:

$$z_i = f(x_i, y_i, \boldsymbol{a}) + \varepsilon_i, \quad i = 1, \ldots, n, \tag{8}$$

where z_i is the result (VGO kinematic viscosity), x_i (average boiling point), and y_i (specific gravity) is the input data; the unknown parameter $\boldsymbol{a} = (a_1, a_2, a_3, a_4, a_5)^T$ is a 5-dimensional vector; and ε_i are random numbers, n = 41; and

$$f(x, y, \boldsymbol{a}) = \exp(\exp(a_1 x^{a_2} y^{a_3} + a_4)) - a_5. \tag{9}$$

To estimate the components of parameter a we used four optimization methods:
Method 1: Classical least squares method:

$$min\left\{F_1(\boldsymbol{a}) = \sum_{i=1}^{n}(z_i - f(x_i, y_i, \boldsymbol{a}))^2 : a \in R^5\right\}. \tag{10}$$

Method 2: Minimization of the sum of absolute values:

$$min\left\{F_2(\boldsymbol{a}) = \sum_{i=1}^{n}|z_i - f(x_i, y_i, \boldsymbol{a})| : a \in R^5\right\}. \tag{11}$$

Method 3: Minimization the sum of squared relative errors:

$$min\left\{F_3(\boldsymbol{a}) = \sum_{i=1}^{n}\left(\frac{z_i - f(x_i, y_i, \boldsymbol{a})}{z_i}\right)^2 : a \in R^5\right\}. \tag{12}$$

Method 4: Minimization the sum of absolute relative errors:

$$min\left\{F_4(\boldsymbol{a}) = \sum_{i=1}^{n}\left|\frac{z_i - f(x_i, y_i, \boldsymbol{a})}{z_i}\right| : a \in R^5\right\}. \tag{13}$$

2.2.2. Computational Minimization

In many cases, there are well-known specialized algorithms for global optimization. Such a case is when f is a monotone function, see for example [25,26]. On the other hand, there are many examples when the sum of squares can have several local minima, see

for example [26] and references therein. In our case, we did not have any conditions guaranteeing the convergence of an iterative process to the global extremum. In this study, one of the goals was to examine that the above-stated four methods are adequate mathematical models capable of satisfactorily describing the data. In order to do this, a minimum of the difference between measured and predicted oil viscosity (different for each model) was searched and sensitivity of model parameters with respect to the data was performed.

As an initial guess, the following modification of Aboul-Seoud and Moharam correction to Walther's model was used:

$$a_1 = 0, a_2 = 0.2, a_3 = 0.2, a_4 = -1, a_5 = -0.8$$

If one starts the computations with the above-mentioned initial conditions, that is, when a_1= 4.3414 and a_1= −15.01620372 many overflow warnings/errors are obtained.

Using a set of quasi-random points in a five-dimensional parametric space in the neighborhood of the initial guess and calculating the values of corresponding criterion function F_j, the computations started (for method 1) with the initial condition:

$$a_1 = 0, a_2 = 1.0889, a_3 = 0.825, a_4 = -1.6, a_5 = -1.6333.$$

More precisely, Halton sequences of quasi-random numbers, with base 2–6 were used to cover the hypercube neighborhood of the initial guess and the lengths of vertices 2. As examples, Halton squares of 20 × 20 points with bases 2, 3, and 4, 5 are plotted on Figure 1. One may compute the initial condition using initial guess and Halton points with indices and bases (0, 2), (8, 3), (10, 4), (3, 5), and (5, 6), respectively.

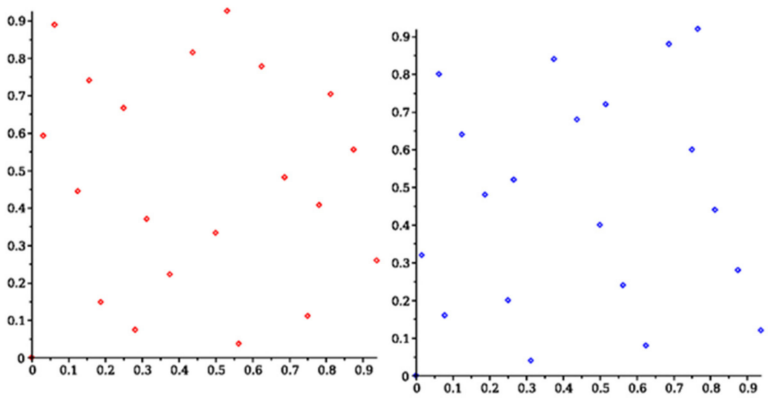


Figure 1. (**a**) Halton square with bases 2, 3. (**b**) Halton square with bases 4, 5.

The discovery strategy for initial conditions of the other three methods was the same.

All computations were performed by the use of CAS Maple and NLPSolve with Modified Newton Iterative Method starting from the corresponding initial condition. The stop-criteria is the absolute difference of two consecutive iterations to be less or equal to 0.01.

2.2.3. Sensitivity Analysis with Respect to Obtained Model Parameters

After successive realization of Newton iterative procedure for method 1, one may receive the following parameters $\widetilde{a}_1 = 0.0000972, \widetilde{a}_2 = 1.5542645, \widetilde{a}_3 = 1.0946136$, $\widetilde{a}_4 = -1.5265719$, $\widetilde{a}_5 = -1.4404829$.

Here, it is worth denoting that the derivatives of $F_1(a) = \sum_{i=1}^{n}(z_i - f(x_i, y_i, a))^2$ were huge numbers outside a "really small" neighborhood of the minimum. Indeed, one may check $F_1(0.0000972, \widetilde{a}_2, \widetilde{a}_3, \widetilde{a}_4, \widetilde{a}_5) = 394.358$ and$F_1(0.0000970, \widetilde{a}_2, \widetilde{a}_3, \widetilde{a}_4, \widetilde{a}_5) = 659.175$.

Therefore, it is necessary to use numbers with at least seven digits after the decimal sign. In Figure 2, the graph of function $F_1(a, \widetilde{a}_2, \widetilde{a}_3, \widetilde{a}_4, \widetilde{a}_5)$ is plotted in blue.

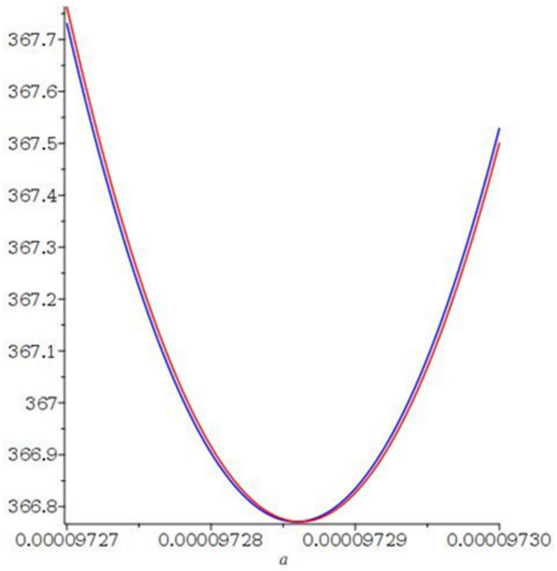


Figure 2. Graphs of functions $F_1(a, \widetilde{a}_2, \widetilde{a}_3, \widetilde{a}_4, \widetilde{a}_5)$ (blue) and $F_1(a, a_2^0, a_3^0, a_4^0, a_5^0)$ (red) in the interval [0.00009727..0.0000973]. Abscissa—variable $\boldsymbol{a}$; Ordinate—variable $\boldsymbol{F_1}$

Moreover, taking in mind the above fact, the appropriateness of the estimated parameters was verified by a sensitivity analysis using perturbations of model parameters in the range of ±20%, similarly as it is described by the authors of [3,4,17,21].

Generating random numbers in the ±20% interval around the obtained values, we were lucky to refine it $a_1^0 = 0.0000973, a_2^0 = 1.5542641,\ a_3^0 = 1.0946132, a_4^0 = -1.5265719,$ $a_5^0 = -1.4404824$. Here $F_1(a_1^0, a_2^0, a_3^0, a_4^0,\ a_5^0) = 367.502$. In Figure 2, the graphs of functions $F_1(a, \widetilde{a}_2, \widetilde{a}_3, \widetilde{a}_4, \widetilde{a}_5)$ and $F_1(a, a_2^0, a_3^0, a_4^0, a_5^0)$ are compared.

The same procedures were to methods 2, 3, and 4. All results are summarized in Table 2.

Table 2. Numerically calculated values of parameter $a^0 = \left(a_1^0, a_2^0, a_3^0, a_4^0, a_5^0 \right)^T$.

Coefficient	Least Squares		Least abs. Errors		Squared rel. Errors		Abs. rel. Errors	
	Before SA	**After SA**	**Before SA**	**After SA**	**Before SA**	**After SA**	**Before SA**	**After SA**
a_1^0	0.0000972	0.0000973	0.0888705	0.0888705	9×10^{-7}	9×10^{-7}	0.0841792	0.0841793
a_2^0	1.5542645	1.5542641	0.6573309	0.657331	2.1851235	2.1851235	0.6533058	0.6533059
a_3^0	1.0946136	1.0946132	0.4784847	0.4784848	1.5193787	1.5193787	0.5075231	0.5075231
a_4^0	−1.5265719	−1.5265719	−5.5717615	−5.571762	−0.4953817	−0.4953818	−5.0323918	−5.0323919
a_5^0	−1.4404829	−1.4404824	−2.4403382	−2.440338	1.9089183	1.9089184	0.0382231	0.0382233

2.2.4. Sensitivity Analysis with Respect to Given Data

Following Ref. [20], the optimization criterion in the four methods were rewritten as constrained optimization problems

$$\min\{F_j(\boldsymbol{a}) : \boldsymbol{a} \in R^p\}, \tag{14}$$

subject to

$$g_i(\boldsymbol{a}) = 0, \quad i = 1, \ldots, n, \tag{15}$$

$$h_i(\boldsymbol{a}) \leq 0, \quad i = 1, \ldots, m. \tag{16}$$

The Lagrangian function for the primal problem (14)–(16) is

$$L(\boldsymbol{a}, \boldsymbol{\lambda}, \boldsymbol{\mu}) = F_j(\boldsymbol{a}) + \sum_{i=1}^{n} \lambda_i g_i(\boldsymbol{a}) + \sum_{i=1}^{m} \mu_i h_i(\boldsymbol{a}), \tag{17}$$

where λ_i are Lagrange multipliers associated with g_i $\boldsymbol{\lambda} = (\lambda_1, \ldots, \lambda_n)$; μ_i are Lagrange multipliers associated with h_i, $\mu = (\mu_1, \ldots, \mu_n)$ The Lagrange dual function is defined by $\widetilde{L}(\boldsymbol{a}, \boldsymbol{\lambda}, \boldsymbol{\mu}) = \inf\{L(\boldsymbol{a}, \boldsymbol{\lambda}, \boldsymbol{\mu}) : a \in R^p\}$. As an infimum of affine functions, the Lagrange dual function is concave. Let us recall that in the local minimum a^0 the necessary conditions, described in Karush-Kuhn-Tucker theorem, are satisfied.

The gradients of Lagrangians of stated methods are calculated. Calculating the arithmetic mean μ_x and deviation σ_x of derivatives with respect to x_i (for example), the standardized deviation of derivatives

$$S_{x_i} = \frac{\frac{\partial L_j(\boldsymbol{a}^0)}{\partial x_i} - \mu_x}{\sigma_x}, \ \ i = 1, \ldots, n, \ j = 1, \ldots, 4, \tag{18}$$

are interpreted as sensitivity coefficients with respect to x_i.

2.2.5. Sensitivity Analysis of Least Squares Method

The classical least square problem is equivalent to the following Lagrange problem

$$\min \sum_{i=1}^{n} \varepsilon_i^2, \tag{19}$$

subject to

$$z_i - f(x_i, y_i, a) = \varepsilon_i, \ \ i = 1, \ldots, n. \tag{20}$$

The Lagrangian function for the least square method (19), (20) is

$$L_1(\boldsymbol{a}) = \sum_{i=1}^{n} (z_i - f(x_i, y_i, \boldsymbol{a}))^2 = \sum_{i=1}^{n} \left(z_i - \exp\left(\exp\left(a_1 x_i^{a_2} y_i^{a_3} + a_4\right)\right) + a_5\right)^2 \tag{21}$$

Therefore, the sensitivities with respect to z_i are

$$\frac{\partial L_1(a^0)}{\partial z_i} = 2\left(z_i - \exp\left(\exp\left(a_1^0 x_i^{a_2^0} y_i^{a_3^0} + a_4^0\right)\right) + a_5^0\right) = 2\left(z_i - f\left(x_i, y_i, \boldsymbol{a}^0\right)\right), \ \ i = 1, \ldots, n. \tag{22}$$

Let

$$\mu_z = \frac{1}{n} \sum_{i=1}^{n} \frac{\partial L_1(\boldsymbol{a}^0)}{\partial z_i} = \frac{2}{n} \sum_{i=1}^{n} z_i - \frac{2}{n} \sum_{i=1}^{n} f\left(x_i, y_i, \boldsymbol{a}^0\right) \tag{23}$$

be the arithmetic mean of derivatives. Let

$$\sigma_z^2 = \frac{1}{n-1} \sum_{i=1}^{n} \left(\frac{\partial L_1(\boldsymbol{a}^0)}{\partial z_i} - \mu_z \right)^2 \tag{24}$$

be the variance of derivatives (here the Bessel's correction is used).

Standardizing, the sensitivity coefficients with respect to z_i are obtained.

$$S_{z_i} = \frac{\frac{\partial L_1(a^0)}{\partial z_i} - \mu_z}{\sigma_z}, \ i = 1, \ldots, n. \tag{25}$$

Similarly, the sensitivities with respect to x_i and y_i are

$$\begin{aligned}\frac{\partial L_1(\boldsymbol{a})}{\partial x_i} &= -2a_1a_2x_i^{a_2-1}y_i^{a_3}exp\big(a_1x_i^{a_2}y_i^{a_3}+exp\big(a_1x_i^{a_2}y_i^{a_3}+a_4\big)+a_4\big)\\ &\quad\times\big(z_i-exp\big(exp\big(a_1x_i^{a_2}y_i^{a_3}+a_4\big)\big)+a_5\big)\\ &= -2a_1a_2x_i^{a_2-1}y_i^{a_3}ln(f(x_i,y_i)+a_5)(f(x_i,y_i)+a_5)(z_i-f(x_i,y_i)),\end{aligned} \tag{26}$$

$$\begin{aligned}\frac{\partial L_1(\boldsymbol{a})}{\partial y_i} &= -2a_1a_3x_i^{a_2}y_i^{a_3-1}exp\big(a_1x^{a_2}y_i^{a_3}+exp\big(a_1x_i^{a_2}y_i^{a_3}+a_4\big)+a_4\big)\\ &\quad\times\big(z_i-exp\big(exp\big(a_1x_i^{a_2}y_i^{a_3}+a_4\big)\big)+a_5\big)\\ &= -2a_1a_3x_i^{a_2}y_i^{a_3-1}ln(f(x_i,y_i)+a_5)(f(x_i,y_i)+a_5)(z_i-f(x_i,y_i)).\end{aligned} \tag{27}$$

Using both equalities in (26) and (27), is derived

$$\frac{\partial L_1(\boldsymbol{a})}{\partial y_i}=\frac{a_3}{a_2}\frac{x_i}{y_i}\frac{\partial L_1(\boldsymbol{a})}{\partial x_i},\ \ i=1,\ldots,n. \tag{28}$$

From (26), arithmetic mean, and variance

$$\mu_x=\frac{1}{n}\sum_{i=1}^{n}\frac{\partial L_1(\boldsymbol{a}^0)}{\partial x_i},\ \sigma_x^2=\frac{1}{n-1}\sum_{i=1}^{n}\left(\frac{\partial L_1(\boldsymbol{a}^0)}{\partial x_i}-\mu_x\right)^2,$$

the sensitivity coefficients with respect to x_i are calculated

$$S_{x_i}=\frac{\frac{\partial L_1(\boldsymbol{a}^0)}{\partial x_i}-\mu_x}{\sigma_x},\ \ i=1,\ldots,n. \tag{29}$$

Analogously, using (28), calculated values of $\frac{\partial L_1(\boldsymbol{a})}{\partial x_i}$ corresponding arithmetic mean μ_y and variance σ_y^2, the sensitivity coefficients with respect to y_i are obtained.

$$S_{y_i}=\frac{\frac{\partial L_1(\boldsymbol{a}^0)}{\partial y_i}-\mu_y}{\sigma_y},\ i=1,\ldots,n. \tag{30}$$

Let us mark, that sometimes it is suitable to have the expressions for derivatives of $\boldsymbol{L}_1$ in terms of Lagrange multipliers λ_i:

$$L_1(\boldsymbol{a})=\sum_{i=1}^{n}\varepsilon_i^2+\sum_{i=1}^{n}\lambda_i(z_i-f(x_i,y_i,\boldsymbol{a})-\varepsilon_i). \tag{31}$$

It is straightforward

$$\begin{aligned}&\frac{\partial L_1(\boldsymbol{a})}{\partial z_i}=\lambda_i=-2\varepsilon_i,\\ &\frac{\partial L_1(\boldsymbol{a})}{\partial x_i}=-\lambda_i\frac{\partial f(x_i,y_i,\boldsymbol{a})}{\partial x_i},\qquad i=1,\ldots,n.\\ &\frac{\partial L_1(\boldsymbol{a})}{\partial y_i}=-\lambda_i\frac{\partial f(x_i,y_i,\boldsymbol{a})}{\partial y_i},\end{aligned} \tag{32}$$

2.2.6. Sensitivity Analysis of Absolute Value Minimization Problem

Analogously, it is suitable to consider the following constrained analog to absolute value minimization problem in method 2:

$$\min\sum_{i=1}^{n}\varepsilon_i, \tag{33}$$

subject to

$$z_i-f(x_i,y_i,\boldsymbol{a})\le\varepsilon_i,\ \ i=1,\ldots,n, \tag{34}$$

$$f(x_i,y_i,\boldsymbol{a})-z_i\le\varepsilon_i,\ \ i=1,\ldots,n, \tag{35}$$

$$0\le\varepsilon_i,\ \ i=1,\ldots,n. \tag{36}$$

The Lagrangian for the problem (33)–(36)

$$L_2(\boldsymbol{a}) = \sum_{i=1}^{n} \varepsilon_i + \sum_{i=1}^{n} \mu_{1i}(z_i - f(x_i, y_i, \boldsymbol{a}) - \varepsilon_i) + \sum_{i=1}^{n} \mu_{2i}(f(x_i, y_i, a) - z_i - \varepsilon_i) + \sum_{i=1}^{n} \mu_{3i}\varepsilon_i,$$

where μ_{ji} ji are the Lagrange multipliers,

$$i = 1, \ldots, n, \; j = 1, 2, 3 \tag{37}$$

Thus

$$\frac{\partial L_2(\boldsymbol{a})}{\partial z_i} = \mu_{1i} - \mu_{2i}, \; i = 1, \ldots, n, \tag{38}$$

$$\frac{\partial L_2(\boldsymbol{a})}{\partial x_i} = a_1 a_2 x_i^{a_2 - 1} y_i^{a_3} (\mu_{2i} - \mu_{1i})(f(x_i, y_i, \boldsymbol{a}) + a_5) \ln(f(x_i, y_i, \boldsymbol{a}) + a_5), \tag{39}$$

$$\frac{\partial L_2(\boldsymbol{a})}{\partial y_i} = a_1 a_3 x_i^{a_2} y_i^{a_3 - 1} (\mu_{2i} - \mu_{1i})(f(x_i, y_i, \boldsymbol{a}) + a_5) \ln(f(x_i, y_i, \boldsymbol{a}) + a_5). \tag{40}$$

It follows from a well-known lemma from the proof of Karush-Kuhn-Tucker conditions (in fact Fritz John conditions), see [27] that if $\boldsymbol{a}^0$ is an optimal solution of the problem (33)–(36), then there exist multipliers μ_0^0, μ_{ji}^0 such that $\mu_0^0, \mu_{ji}^0 \geq 0$, $j = 1, 2, 3$, $i = 1, \ldots, n$, not all zero, and

$$\mu_0^0 \nabla_{\varepsilon} \sum_{i=1}^{n} \varepsilon_i + \sum_{i \in J_1(a^0)} \mu_{1i}^0 \nabla_e (z_i - f(x_i, y_i, a) - \varepsilon_i) + \sum_{i \in J_2(a^0)} \mu_{2i}^0 \nabla_{\varepsilon} (f(x_i, y_i, a) - z_i - \varepsilon_i) + \sum_{i \in J_3(a^0)} \mu_{3i}^0 \nabla_{\varepsilon} (-\varepsilon_i) = 0, \tag{41}$$

where:

$$J_1\left(a^0\right) = \{i \in 1, \ldots, n : z_i - f(x_i, y_i, a) - \varepsilon_i = 0\}, \; J_2\left(a^0\right) = \{i \in 1, \ldots, n : f(x_i, y_i, a) - z_i - \varepsilon_i = 0\}$$

$$J_3\left(a^0\right) = \{i \in 1, \ldots, n : \varepsilon_i = 0\}$$

are the corresponding active conditions, $\varepsilon = (\varepsilon_1, \varepsilon_2, \ldots, \varepsilon_n)^T$ Simplifying

$$\mu_0^0 \boldsymbol{e} - \sum_{i \in J_1(a^0)} \mu_{1i}^0 \boldsymbol{e}_i - \sum_{i \in J_2(a^0)} \mu_{2i}^0 \boldsymbol{e}_i - \sum_{i \in J_3(a^0)} \mu_{3i}^0 \boldsymbol{e}_i = 0, \tag{42}$$

where $\boldsymbol{e}_i$ is the i-th unit vector and $\boldsymbol{e} = (1, \ldots, 1)^T$. Let us note:

$$J_1\left(a^0\right) \cup J_2\left(a^0\right) = \{1, 2, \ldots, n\} = J_3\left(a^0\right) \text{ and } J_1\left(a^0\right) \cap J_2\left(a^0\right) = \varnothing. \tag{43}$$

Hence, one may construct a non-negative solution of the linear system setting:

$$\mu_{1i}^0 = 0, \mu_{2i}^0 = 1 \text{ if } \varepsilon_i < 0, \text{ i.e., } i \in J_2, \tag{44}$$

$$\mu_{1i}^0 = 1, \mu_{2i}^0 = 0 \text{ if } \varepsilon_i > 0, \text{i.e., } i \in J_1, \tag{45}$$

$$\mu_0^0 = 1, \mu_{3i} = 0, \; i = 1, \ldots, n. \tag{46}$$

2.2.7. Sensitivity Analysis of Squared Relative Errors

Analogously to previous subsections, the minimization problem in Method 3 is equivalent to the following Lagrange problem. The equivalent Lagrange problem is

$$\min \sum_{i=1}^{n} \varepsilon_i^2, \tag{47}$$

subject to

$$z_i - (f(x_i, y_i, a)) = z_i \varepsilon_i,\ i = 1, \ldots, n. \tag{48}$$

The Lagrangian is

$$L_3(a) = \sum_{i=1}^{n} \left(\frac{z_i - f(x_i, y_i, a)}{z_i} \right)^2 = \sum_{i=1}^{n} \left(1 - \frac{exp\left(exp\left(a_1 x_i^{a_2} y_i^{a_3} + a_4\right)\right) - a_5}{z_i} \right)^2 \tag{49}$$

The first derivatives are (here the already calculated derivatives of L_1 are used)

$$\frac{\partial L_3(a^0)}{\partial z_i} = \frac{1}{z_i^2} \frac{\partial L_1(a^0)}{\partial z_i} - \frac{1}{2z_i^3} \left(\frac{\partial L_1(a^0)}{\partial z_i} \right)^2, \tag{50}$$

$$\frac{\partial L_3(a^0)}{\partial x_i} = \frac{1}{z_1^2} \frac{\partial L_1(a^0)}{\partial x_i}, \tag{51}$$

$$\frac{\partial L_3(a^0)}{\partial y_i} = \frac{1}{z_1^2} \frac{\partial L_1(a^0)}{\partial y_i}, \tag{52}$$

where $i = 1, \ldots, n$.

The formulas for sensitivity analysis of the sum of absolute relative errors are omitted because they are similar to the explanation in Section 2.2.7.

3. Results

The data in Table 1 indicate that the selected vacuum gas oils (VGO) differentiate significantly in their properties. The most important for modeling viscosity oil properties: specific gravity, and average boiling point [14] varied in the range 0.838 ÷ 1.177 for specific gravity, and 309 ÷ 488 °C for average boiling point. The VGO viscosity at 80 °C varied between 3.6 and 312.8 mm^2/s.

The Bayesian approach was used over several classical distributions to find the distribution functions of specific gravity (SG) and average boiling point (ABP). Using the Bayesian information criterion, one may conclude that the best distribution for SG is the normal distribution with mean and variance 0.98712, 0.0771899, respectively. The second and third candidates for continuous probability distribution are Gamma distribution and LogNormal distribution. The histogram and PDF (Probability Density Function) of SG data are plotted on Figure 3.

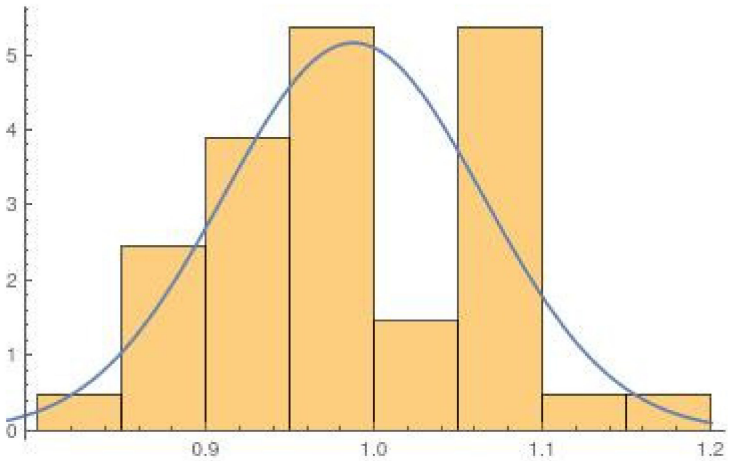


Figure 3. Normal distribution plot for the SG data of the VGOs from Table 1.

For the second data—ABP, using similar arguments, we again obtained the normal distribution with mean and variance 416.284, 39.0181, respectively. The histogram and PDF function are plotted in Figure 4.

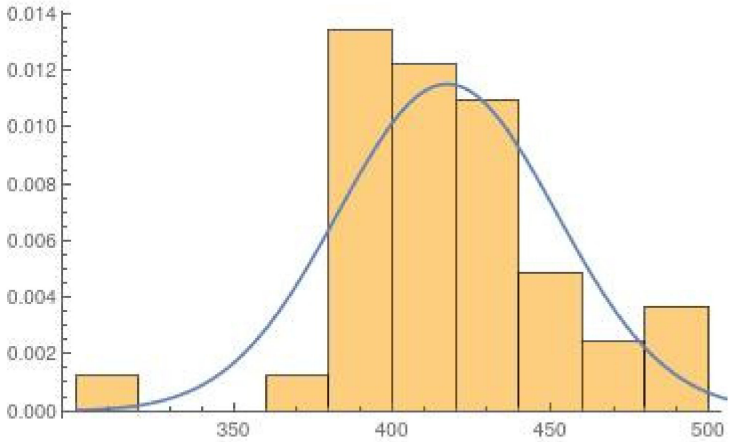


Figure 4. Normal distribution plot for the ABP data of the VGOs from Table 1.

Table 2 presents data about regression coefficients for the four methods obtained after application of the Newton iterative procedure, and after the sensitivity analysis with respect to obtained parameters. These data show that the performed sensitivity analysis with respect to the model parameters in most cases led to a modification of the values of the regression coefficients.

Table 3 indicates data of calculated viscosity of the 41 VGOs from Table 1, error, absolute relative error, and average abs. rel. error (AARE), also known as *%AAD* (average absolute deviation) by the use of the optimized values of the regression coefficients from Table 2 (model parameters after sensitivity analysis). The errors, and the *%AAD* were computed as shown in Equations (53) and (54) respectively:

$$\text{Error } (E): \ E = \left(\frac{v_{exp} - v_{calc}}{v_{exp}} \right) \times 100 \tag{53}$$

$$\%AAD = \frac{1}{n} \sum_{i=1}^{n} \frac{|v_{exp} - v_{calc}|}{v_{exp}} \times 100 \tag{54}$$

Considering the *%AAD* as a criterion for classification of the four studied methods the method *%AAD* increases in the order: Method 3 < Method 4 < Method 1 < Method 2.

Table 4 shows the standardized sensitivities for the four studied estimation methods.

Table 3. Calculated results for four estimation methods, calc.—calculated value; rel.error.—relative error (in %).

		Least Squares (Method 1)			**Least abs. Errors (Method 2)**			**Squared rel. Errors (Method 3)**			**Abs. rel. Errors (Method 4)**		
Nr		**calc.**	**Error**	**rel. Error**	**calc.**	**Error**	**rel. Error**	**calc.**	**Error**	**rel. Error**	**calc.**	**Error**	**rel. Error**
1	HAGO-1	9.79	−2.52	34.7	9.74	−2.47	33.9	8.48	−1.21	16.7	7.99	−0.72	10
2	LVGO-1	13.25	−1.17	9.7	13.09	−1.01	8.4	12.34	−0.26	2.1	11.6	0.48	4
3	HVGO-1	50.96	−1.05	2.1	49.76	0.15	0.3	51.55	−1.64	3.3	47.02	2.89	5.8
4	HAGO-2	9.98	3.62	26.6	9.93	3.67	27	8.69	4.91	36.1	8.22	5.38	39.5
5	LVGO-2	13.33	1.87	12.3	13.21	1.99	13.1	12.41	2.79	18.3	11.8	3.4	22.4
6	HVGO-2	64.42	−2.32	3.7	63.15	−1.05	1.7	64.75	−2.65	4.3	60.14	1.96	3.2
7	HAGO-3	8.25	4.65	36.1	8.27	4.63	35.9	6.71	6.19	48	6.43	6.47	50.1
8	LVGO-3	11.22	5.48	32.8	11.14	5.56	33.3	10.08	6.62	39.7	9.58	7.12	42.6
9	HVGO-3	38.5	−3.7	10.6	37.9	−3.1	8.9	38.86	−4.06	11.7	36.37	−1.57	4.5
10	FCC SLO-1	5.72	−2.16	60.8	5.95	−2.39	67.2	3.72	−0.16	4.5	3.93	−0.37	10.3

Table 3. *Cont.*

Nr		Least Squares (Method 1)			Least abs. Errors (Method 2)			Squared rel. Errors (Method 3)			Abs. rel. Errors (Method 4)		
		calc.	Error	rel. Error	calc.	Error	rel. Error	calc.	Error	rel. Error	calc.	Error	rel. Error
11	FCC SLO-2	13.72	−3.82	38.5	13.77	−3.87	39.1	12.76	−2.86	28.9	12.8	−2.9	29.3
12	FCC SLO-3	17.82	−1.62	10	17.89	−1.69	10.4	17.19	−0.99	6.1	17.27	−1.07	6.6
13	FCC SLO-4	21.53	−0.23	1.1	21.68	−0.38	1.8	21.1	0.2	0.9	21.43	−0.13	0.6
14	FCC SLO-5	17.88	−0.48	2.8	17.97	−0.57	3.3	17.25	0.15	0.9	17.41	−0.01	0
15	FCC SLO-6	28.27	5.49	16.3	28.74	5.02	14.9	28.01	5.75	17	29.34	4.42	13.1
16	FCC SLO-7	23.89	0.32	1.3	24.15	0.06	0.2	23.54	0.67	2.8	24.21	0	0
17	FCC SLO-8	18.38	0.09	0.5	18.48	−0.01	0	17.78	0.69	3.8	17.96	0.51	2.8
18	FCC SLO-9	23.7	4.81	16.9	23.99	4.52	15.9	23.33	5.18	18.2	24.11	4.4	15.4
19	FCC SLO-10	312.7	0.1	0	312.8	0	0	288.07	24.73	7.9	316.69	−3.89	1.2
20	FCC SLO-11	27.18	−5.94	27.9	27.66	−6.42	30.2	26.87	−5.63	26.5	28.3	−7.06	33.2
21	VGO blend	13.82	0.37	2.6	13.49	0.7	5	13.04	1.15	8.1	11.68	2.51	17.7
22	HAGO-4	9.81	−2.41	32.6	9.68	−2.28	30.8	8.54	−1.14	15.4	7.77	−0.37	5.1
23	LVGO-4	10.25	−2.65	34.8	10.1	−2.5	32.9	9.03	−1.43	18.8	8.25	−0.65	8.5
24	HVGO-4	23.44	8.16	25.8	22.67	8.93	28.3	23.48	8.12	25.7	20.59	11.01	34.8
25	HAGO-5	10.69	2.31	17.7	10.63	2.37	18.2	9.49	3.51	27	9.01	3.99	30.7
26	LVGO-5	11.02	1.98	15.3	10.97	2.03	15.6	9.84	3.16	24.3	9.43	3.57	27.5
27	HVGO-5	54.1	3.4	5.9	53.38	4.12	7.2	54.46	3.04	5.3	51.49	6.01	10.4
28	FCC SLO-12	26.07	−3.87	17.4	26.49	−4.29	19.3	25.74	−3.54	15.9	26.98	−4.78	21.5
29	VBGO-1	14.48	0.22	1.5	14.19	0.51	3.5	13.75	0.95	6.5	12.53	2.17	14.8
30	VBGO-2	13.44	0.06	0.5	13.2	0.3	2.2	12.58	0.92	6.8	11.56	1.94	14.4
31	FCC SLO-13	11.5	3	20.7	11.56	2.94	20.3	10.32	4.18	28.8	10.35	4.15	28.6
32	FCC SLO-14	18.34	−2.14	13.2	18.5	−2.3	14.2	17.71	−1.51	9.3	18.12	−1.92	11.9
33	HTVGO-1	10.37	0.03	0.3	10.19	0.21	2	9.19	1.21	11.7	8.27	2.13	20.5
34	HTVGO-2	9.86	−0.26	2.7	9.7	−0.1	1.1	8.6	1	10.4	7.76	1.84	19.2
35	BG LIGHT	6.19	−2.49	67.4	6.32	−2.62	70.8	4.32	−0.62	16.7	4.2	−0.5	13.5
36	PEMBINA	9.88	−2.08	26.7	9.73	−1.93	24.7	8.62	−0.82	10.5	7.8	0	0
37	EKOFISK	11.8	−4	51.3	11.55	−3.75	48.1	10.8	−3	38.5	9.67	−1.87	24
38	BRENT	8.28	0.12	1.4	8.25	0.15	1.8	6.78	1.62	19.3	6.28	2.12	25.3
39	BOW RIVER	11.23	−1.73	18.2	11.07	−1.57	16.5	10.13	−0.63	6.6	9.31	0.19	2
40	COKER	19.68	1.02	4.9	19.53	1.17	5.7	19.26	1.44	7	18.52	2.18	10.5
41	BU ATTIFEL	8.75	−0.45	5.4	8.61	−0.31	3.7	7.35	0.95	11.4	6.5	1.8	21.7
AARE (%AAD)				17.3			17.5			15.2			16.0

Table 4. Standardized sensitivities for four estimation methods.

Nr.	Least Squares			Least abs. Errors			Squared rel. Errors			Abs. rel. Errors		
	S_{z_i}	S_{x_i}	S_{y_i}	S_{z_i}	S_{x_i}	S_{y_i}	S_{z_i}	S_{x_i}	S_{y_i}	S_{z_i}	S_{x_i}	S_{y_i}
1	−0.83	0.17	0.17	−0.96	0.2	0.21	−1.28	0.93	0.95	−1.34	0.8	0.87
2	−0.39	0.13	0.13	−0.96	0.24	0.25	0.06	0.12	0.12	0.94	−0.86	−0.86
3	−0.35	0.85	0.93	1.01	−0.43	−0.57	0.13	0.29	0.32	0.34	−1.27	−1.41
4	1.2	−0.25	−0.25	1.01	0.1	0.1	1.07	−1.12	−1.12	0.6	−0.47	−0.45
5	0.62	−0.22	−0.21	1.01	0.07	0.06	0.69	−0.82	−0.82	0.66	−0.71	−0.68
6	−0.77	**2.59**	**2.75**	−0.96	0.96	1.13	0.12	0.41	0.44	0.31	−1.41	−1.52
7	1.54	−0.23	−0.22	1.01	0.12	0.12	1.2	−1.11	−1.1	0.54	−0.35	−0.32
8	1.81	−0.47	−0.46	1.01	0.09	0.08	0.93	−1.23	−1.22	0.5	−0.49	−0.45
9	−1.22	**2.07**	**2.17**	−0.96	0.56	0.64	−0.04	1.02	1.1	−0.14	1.3	1.46
10	−0.71	0.05	0.04	−0.96	0.17	0.17	−0.55	0.18	0.16	**−2.92**	0.57	0.52
11	−1.26	0.49	0.42	−0.96	0.25	0.25	−1.86	**2.17**	1.94	−1.14	1.23	1.16
12	−0.54	0.31	0.28	−0.96	0.3	0.3	−0.05	0.41	0.38	−0.5	1.12	1.08
13	−0.08	0.06	0.05	−0.96	0.35	0.35	0.19	−0.06	−0.06	−0.31	1.14	1.1
14	−0.16	0.09	0.08	−0.96	0.3	0.3	0.19	−0.06	−0.05	−0.41	1.06	1.02
15	1.81	**−2.14**	−1.87	1.01	−0.15	−0.15	0.39	−1.08	−0.96	0.41	−1.13	−1

Table 4. *Cont.*

Nr.	Least Squares			Least abs. Errors			Squared rel. Errors			Abs. rel. Errors		
	S_{z_i}	S_{x_i}	S_{y_i}	S_{z_i}	S_{x_i}	S_{y_i}	S_{z_i}	S_{x_i}	S_{y_i}	S_{z_i}	S_{x_i}	S_{y_i}
16	0.1	−0.1	−0.08	1.01	−0.08	−0.08	0.22	−0.19	−0.17	−0.25	1.19	1.13
17	0.03	−0.02	−0.02	−0.96	0.31	0.31	0.27	−0.24	−0.21	0.68	−1.08	−0.96
18	1.59	−1.45	−1.27	1.01	−0.08	−0.08	0.44	−1.06	−0.96	0.45	−1.04	−0.92
19	0.03	−0.85	−0.74	1.01	**−6.15**	**−6.09**	0.17	−1.06	−0.94	0.12	**2.22**	**2.06**
20	−1.96	**2.19**	1.89	−0.96	0.44	0.43	−0.68	**2.53**	**2.24**	−0.47	1.68	1.55
21	0.12	−0.04	−0.05	1.01	0.07	0.05	0.44	−0.4	−0.45	0.73	−0.71	−0.78
22	−0.8	0.15	0.17	−0.96	0.2	0.21	−1.12	0.82	0.91	−1.25	0.72	0.86
23	−0.87	0.19	0.2	−0.96	0.2	0.21	−1.41	1.05	1.16	−1.26	0.77	0.91
24	**2.7**	**−2.16**	**−2.48**	1.01	−0.04	−0.09	0.49	−1.22	−1.44	0.36	−0.68	−0.78
25	0.76	−0.18	−0.18	1.01	0.1	0.09	0.97	−0.99	−0.98	0.69	−0.57	−0.54
26	0.65	−0.17	−0.16	1.01	0.09	0.09	0.92	−0.94	−0.92	0.71	−0.61	−0.57
27	1.12	**−3.04**	**−3.15**	1.01	−0.5	−0.62	0.21	−0.44	−0.47	0.31	−1.26	−1.33
28	−1.28	1.34	1.16	−0.96	0.42	0.42	−0.28	1.37	1.22	−0.39	1.51	1.4
29	0.07	−0.03	−0.03	1.01	0.06	0.04	0.38	−0.33	−0.36	0.73	−0.76	−0.81
30	0.02	−0.01	−0.01	1.01	0.07	0.05	0.41	−0.34	−0.37	0.79	−0.75	−0.78
31	0.99	−0.29	−0.24	1.01	0.08	0.08	0.92	−1.12	−0.98	0.64	−0.65	−0.55
32	−0.71	0.44	0.38	−0.96	0.31	0.31	−0.18	0.67	0.59	−0.53	1.22	1.14
33	0.01	0	0	1.01	0.1	0.09	0.69	−0.48	−0.55	0.91	−0.59	−0.65
34	−0.09	0.02	0.02	−0.96	0.2	0.21	0.68	−0.43	−0.49	0.99	−0.58	−0.64
35	−0.82	0.06	0.07	−0.96	0.17	0.17	**−2.66**	0.72	0.78	**−2.89**	0.55	0.65
36	−0.69	0.13	0.15	−0.96	0.2	0.21	−0.64	0.53	0.61	−1.12	0.68	0.83
37	−1.32	0.36	0.4	−0.96	0.22	0.23	**−3.5**	**2.64**	**3.02**	−1.42	0.94	1.13
38	0.04	−0.01	−0.01	1.01	0.12	0.11	1.16	−0.67	−0.73	1.04	−0.49	−0.51
39	−0.57	0.14	0.15	−0.96	0.21	0.22	−0.24	0.35	0.38	1.18	−0.78	−0.82
40	0.34	−0.22	−0.22	1.01	−0.01	−0.03	0.33	−0.42	−0.42	0.59	−0.96	−0.94
41	−0.15	0.02	0.03	−0.96	0.19	0.2	0.81	−0.42	−0.52	1.09	−0.5	−0.59

Note: The bold figures mean high values of standardized sensitivities.

The data in Table 4 display that Method 1 VGO under numbers 6, 9, 15, 20, 24, 27 exhibited a high sensitivity for the data of ABP (x). The VGO under number 24 demonstrated high sensitivity for the data of viscosity (z). The VGOs under numbers 6, 9, 24, 27 indicate a high sensitivity for the data of SG (y). For Method 2 only the data of VGO under number 19 exhibits a high sensitivity for the data of ABP (x) and SG (y). For Method 3, the data of VGO under numbers 35 and 37 demonstrate a high sensitivity for the data of viscosity (z). The VGOs under numbers 11, 20, and 37 show high sensitivity for the data of ABP (x). The data for SG (y) of VGOs under numbers 20, and 37 display a high sensitivity. Method 4 indicates a high sensitivity for VGOs under numbers 10 and 35 for the data of viscosity (z), under number 19 for ABP (x) and SG (y).

Table 5 presents data about the means and standard deviations of derivatives. It is evident from these data that Method 3 is characterized with the lowest standard deviation of derivatives followed by Method 4. Methods 1 and 2 have two and three orders of magnitude higher standard deviation of derivatives than those of Methods 3 and 4.

Akaike Information Criterion.

Table 5. Means and standard deviations of derivatives.

	Least Squares		Least abs. Errors		Squared rel. Errors		Absl. rel. Errors	
	μ	σ	μ	σ	μ	σ	μ	σ
with respect to z_i	0	6.06	−0.02	−0.02	0	0.04	−0.01	0.1
with respect to x_i	0	2.81	−0.28	−0.28	0	0	0	0.02
with respect to y_i	0.16	**1409.28**	**−129.95**	**848.78**	0	2.54	−0.16	8.36

Table 6 presents independent data (kin. viscosity at 80 °C; ABP, and SG)) for 43 gas oils to verify the capability of the four methods to predict viscosity. These data include gas oils ranging from light gas oil to VGO. The SG and ABP for this independent data set vary between 0.805 and 1.006, and between 205 and 463 °C, respectively. The kinematic viscosity varies between 0.8 and 28.1 mm2/s. The % AAD increases in the order Method 3 (18.2%) < Method 4 (28.3%) < Method 1 (61.8%) < Method 2 (67.8%).

Table 6. Independent data for gas oils (from light gas oil to VGO) to verify the capability of the four methods to predict viscosity at 80 °C.

					Calculated Viscosity, mm^2/s				Abs. Relative Error, %			
Nr	**VGO and Light Gas Oils**	**Kin. vis. at 80 °C, mm^2/s**	**ABP**	**SG**	**Method 1**	**Method 2**	**Method 3**	**Method 4**	**Method 1**	**Method 2**	**Method 3**	**Method 4**
1	HYDRA	9.9	439	0.8861	10.4	10.2	9.2	8.3	5.0	3.0	6.9	16.0
2	EL BUNDUQ	11.6	434	0.9240	12.3	12.0	11.3	10.3	5.6	3.5	2.8	11.1
3	SUNNILAND	13.3	444	0.9420	15.7	15.3	15.0	13.7	17.5	14.9	12.7	3.1
4	Urals	14.4	445	0.9235	14.0	13.6	13.2	11.9	3.1	5.4	8.5	17.1
5	INNES	10.5	435	0.8793	9.7	9.6	8.5	7.7	7.3	8.8	19.4	27.0
6	LOKELE	15.4	441	0.9581	16.7	16.4	16.1	14.9	8.4	6.3	4.7	3.1
7	Cold Lake	8.0	407	0.9291	9.6	9.6	8.3	7.8	20.6	19.5	4.0	2.3
8	CANMET	5.4	376	0.9446	7.9	7.9	6.3	6.1	45.5	46.3	15.8	12.8
9	VISBROKEN	5.0	382	0.9696	9.2	9.2	7.8	7.6	84.3	84.1	56.1	51.3
10	CHAMPION EXPORT	14.0	426	0.9721	15.1	14.9	14.4	13.5	8.1	6.5	2.9	3.2
11	UDANG	9.3	455	0.8460	9.7	9.5	8.5	7.5	4.8	2.4	8.6	19.2
12	KAKAP	4.8	424	0.8570	8.0	7.9	6.5	6.0	66.6	65.4	34.5	24.0
13	DAQUING	8.2	446	0.8651	10.0	9.7	8.7	7.8	21.4	18.9	6.4	4.9
14	SERGIPANO PLATFORMA	9.2	437	0.8715	9.5	9.3	8.2	7.4	3.2	1.5	11.0	19.5
15	LAKE ARTHUR	8.6	420	0.8766	8.4	8.4	7.0	6.4	1.9	2.7	19.0	25.2
16	MARGHAM LIGHT	6.3	415	0.8691	7.9	7.9	6.3	5.9	24.8	24.3	0.1	6.8
17	SYNTHETIC OSA STREAM	9.3	411	0.9434	10.7	10.6	9.6	9.0	15.4	14.1	2.7	3.6
18	COLD LAKE BLEND	28.1	463	0.9655	25.0	24.4	25.0	22.9	11.1	13.1	11.2	18.4
19	DULANG	4.8	409	0.8504	7.0	7.0	5.3	5.0	44.6	45.3	9.3	3.4
20	HARRIET	5.6	422	0.8902	9.1	9.0	7.7	7.1	63.3	61.4	38.6	27.7
21	TIA JUANA P	26.1	461	0.9673	24.4	23.9	24.4	22.5	6.4	8.4	6.6	14.0
22	TIA JUANA M	19.7	450	0.9373	16.2	15.8	15.7	14.2	17.6	19.6	20.5	27.7
23	SOUEDIE	20.3	454	0.9529	19.4	19.0	19.1	17.4	4.3	6.4	6.0	13.9
24	ARAB HEAVY	11.7	450	0.9285	15.3	15.0	14.7	13.3	30.8	27.5	25.2	13.3
25	ARAB MEDIUM	8.2	445	0.9183	13.5	13.2	12.7	11.5	65.4	61.5	55.3	40.4
26	ARAB LIGHT	10.2	449	0.9196	14.3	14.0	13.6	12.2	40.2	36.6	32.9	19.9
27	MAGNUS	13.1	451	0.8995	12.8	12.5	11.9	10.6	2.2	4.7	9.0	18.6
28	GULLFAKS	16.4	453	0.9204	15.1	14.7	14.5	13.0	7.7	10.1	11.7	20.6
29	FLOTTA BLEND	16.4	458	0.9168	15.6	15.2	15.0	13.4	4.6	7.2	8.3	17.9
30	EKOFISK	10.6	444	0.8963	11.7	11.4	10.6	9.6	10.0	7.5	0.4	9.8
31	HT Kerosene	0.8	205	0.8053	3.4	3.9	0.8	1.6	323.5	389.2	1.9	101.8
32	HTDiesel-2	1.2	251	0.8310	3.7	4.2	1.3	1.9	211.2	249.1	5.5	61.0
33	HTDiesel-3	2.1	310	0.8576	4.5	4.8	2.2	2.7	114.0	129.7	6.2	26.2
34	FCC LCO	1.1	250	0.9461	4.2	4.6	1.8	2.4	281.6	317.0	68.1	119.8
35	FCC HCO-1	2.2	309	0.9960	5.9	6.1	3.9	4.2	166.7	176.7	76.9	89.3
36	FCC HCO-2	3.4	325	0.9950	6.4	6.6	4.6	4.7	89.1	94.2	34.1	39.7
37	FCC HCO-3	4.4	340	1.0064	7.4	7.6	5.7	5.8	69.0	71.7	30.4	32.7

Table 6. *Cont.*

Nr	VGO and Light Gas Oils	Kin. vis. at 80 °C, mm²/s	ABP	SG	Calculated Viscosity, mm²/s				Abs. Relative Error, %			
					Method 1	Method 2	Method 3	Method 4	Method 1	Method 2	Method 3	Method 4
38	SRLVGO	2.4	314	0.8800	4.7	5.0	2.5	2.9	97.3	109.9	5.4	20.7
39	SRVGO-1	1.1	246	0.8345	3.7	4.2	1.2	1.9	236.9	278.7	11.8	73.4
40	SRVGO-2	1.37	269	0.8456	3.9	4.4	1.5	2.1	187.3	217.8	11.3	54.9
41	VBGO-3	1.7	295	0.8618	4.3	4.7	2.0	2.5	153.4	174.5	17.4	45.7
42	SRHVGO-1	7.75	442	0.9230	13.3	13.0	12.5	11.3	72.1	68.3	61.1	46.4
43	SRHVGO-1	12.39	440	0.9227	13.1	12.8	12.2	11.1	5.4	3.1	1.7	10.6
%AAD									61.8	67.8	18.2	28.3

3.1. Evaluation of the Accuracy of Viscosity Estimation by the Studied Four Methods

Besides the error (53), and $\%AAD$ (54) the following additional statistical parameters were used to evaluate the accuracy of viscosity estimation by the studied four methods for the data set of Table 1 [3]:

$$\text{Standard error (SE)}: \; SE = \left(\sum \left(\frac{(v_{exp} - v_{calc})^2}{n} \right) \right)^{\frac{1}{2}} \tag{55}$$

$$\text{Relative standard error (RSE)}: \; RSE = \frac{SE}{mean\ of\ the\ sample} \times 100 \tag{56}$$

$$\text{Sum of square errors (SSE)}: \; SSE = \sum \frac{1}{v_{exp}^2} (v_{exp} - v_{calc})^2 \tag{57}$$

$$\text{Residual (R)}: \; R = v_{exp} - v_{calc}, \tag{58}$$

$$\text{Relative Error (RE)}: \; RE = (\sum (\frac{v_{exp} - v_{calc}}{v_{exp}})) \times 100 \tag{59}$$

Table 7 summarizes the statistical analyses for the four studied methods employing the data in Table 1. According to the statistical parameters standard error, relative standard error Methods 1 and 2 surpass in the accuracy of viscosity prediction Methods 3 and 4. However, regarding the statistical parameters relative error, the sum of square errors, $\%AAD$, Method 3 seems to be the best. It is difficult to distinguish the best method on the basis of the statistical parameters estimated by Equations (53)–(59). The Akaike information criterion (AIC) and Bayesian information criterion (BIC) were found capable of estimating the relative quality of a statistical method, and thus being able of providing means for model selection [13,28,29] when several models are available. Below the estimation of AIC and BIC for the four studied methods is summarized:

Table 7. Statistical analysis of the four methods for the data from Table 1.

	Method 1	**Method 2**	**Method 3**	**Method 4**
Min E	−67.3	−70.8	−38.5	−33.3
Max E	36.0	35.9	48.0	50.2
RE	−232.4	−217.0	149.8	296.5
SE	3.1	3.1	5.1	3.7
RSE	12.0	12.2	20.0	14.5
SSE	2.4	2.5	1.5	1.7
%AAD	17.3	17.5	15.2	16.0
R^2	0.996	0.9959	0.9948	0.9953
Slope	0.996	0.9954	0.9244	1.0118
Intercept	0.1023	0.0095	0.5351	−1.6381
AIC	211	175	−14	190
BIC	220	184	−5	198

Consider the obtained errors $\{\epsilon_1, \ldots, \epsilon_n\}$ as independent random samples from a density function $f(\epsilon\_i|\theta)$, $n = 41$. Supposing normal distribution of errors:

$$f(x|\theta) = f(x|\{\mu, \sigma\}) = \frac{1}{\sigma\sqrt{2\pi}} \exp\left(-\frac{1}{2}\left(\frac{x-\mu}{\sigma}\right)^2\right). \tag{60}$$

Then by the definition of likelihood function:

$$L(\theta) = \prod_{i=1}^{n} f(\varepsilon_i|\theta) = \prod_{i=1}^{n} \frac{1}{\sigma\sqrt{2\pi}} \exp\left(-\frac{1}{2}\left(\frac{\varepsilon_i-\mu}{\sigma}\right)^2\right) \tag{61}$$

The function L has maximum, if

$$\mu = \hat{\mu} = \frac{1}{n}\sum_{i=1}^{n} \varepsilon_i \text{ and } \sigma^2 = \hat{\sigma}^2 = \frac{1}{n}\sum_{i=1}^{n} (\varepsilon_i - \hat{\mu})^2. \tag{62}$$

Method 1: Obtained errors: $\{\varepsilon_1, \ldots, \varepsilon_n\}$. Estimating the maximizers of likelihood function: $\hat{\mu} = 9.0452$ and $\sigma = 167.7992$. Hence the Akaike information criterion value is

$$AIC_1 = 2 \times [number\ of\ parameters] - 2 \times \ln\left(L\left(\hat{\theta}\right)\right) \approx 211 \tag{63}$$

Analogously $AIC_2 \approx 175$, $AIC_3 = -14$, and $AIC_4 = 190$.
For model comparison, the model with the lowest *AIC* score is preferred [29].
Bayesian information criterion
The Bayesian information criterion is defined by

$$BIC = [\text{number of parameters}] \times \ln([\text{number of data points}]) - 2 \times \ln\left(L\left(\hat{\theta}\right)\right), \tag{64}$$

In our case:

$$BIC_1 \approx 220, BIC_2 \approx 184, BIC_3 \approx -5, BIC_4 \approx 198.$$

Again: the model with the lowest *BIC* score is preferred.
On the base of *AIC*, and *BIC* one may conclude that Method 3 is the model with the highest quality.

3.2. Sensitivity Analysis with Respect to Given Data

Tables 4 and 5 summarizes the means and standard deviations of derivatives of the four investigated methods.

The variances σ^2 in the datasets of derivatives (especially with respect to y_i, $i = 1, ..., n$, we have $\sigma_y^2 \approx 1408$ *or* 850, respectively) are huge in the first two methods.

In Table 4 the extreme values of sensitive coefficients are marked in bold. In fact, the extreme value of the deviation of the derivative is a sign of a possible problem: the model is not suitable for certain data or the given point does not correspond to the model, etc. On the other hand, different objective functions and corresponding analyses produce different extremal values in the set of all sensitivities. Therefore, it is a good idea to perform sensitivity analysis through different objective/target functions to one and the same mathematical model and to analyze the obtained values in order to improve the model or to exclude an initially given data. In Figure 5 the distributions of sensitivity coefficients in the four methods are presented.

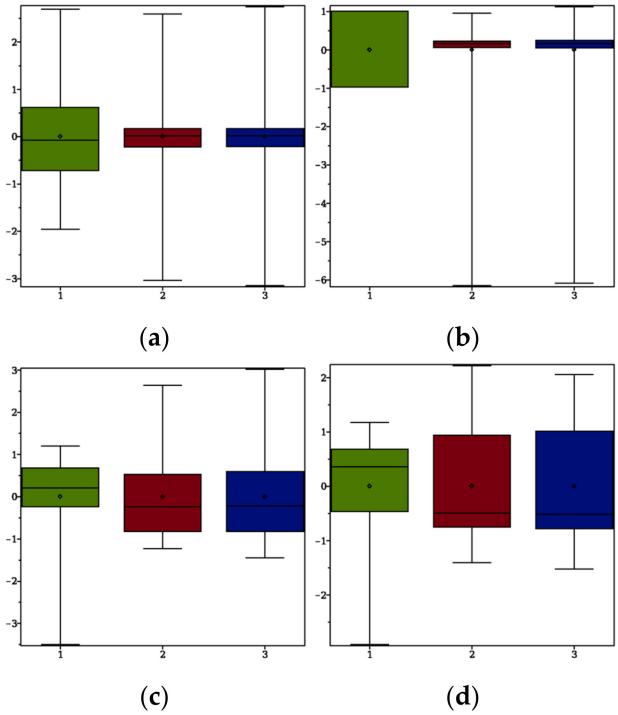
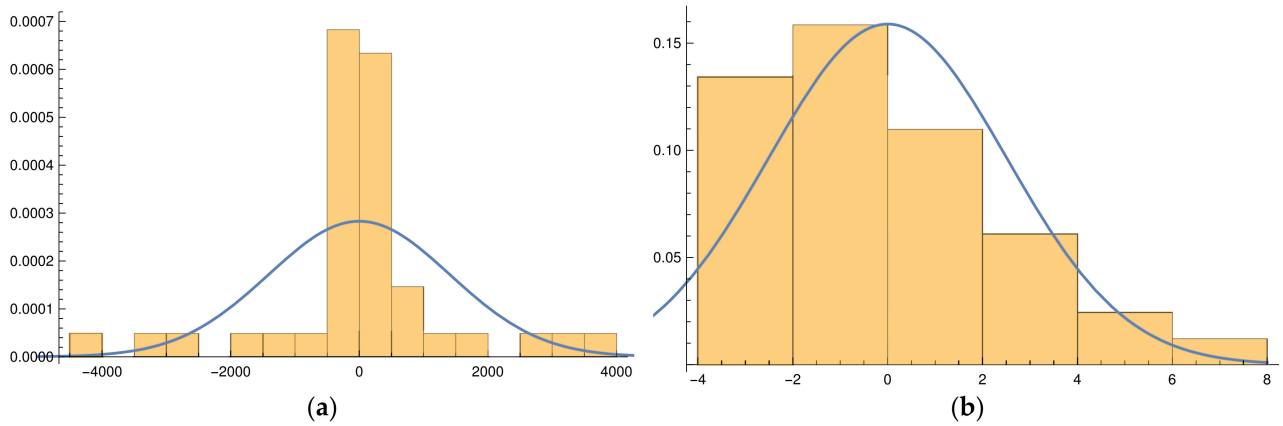


Figure 5. The boxed-graphs of sensitivity coefficients with respect to z, x, and y for all four methods. Each boxed-graph is based on the five-number statistical characteristics; minimum, first quartile, median, third quartile, and maximum (the central rectangle represents from the first quartile to the third quartile; the segment inside the rectangle is the median; the dot is the mean). (**a**)—Method 1; (**b**)—Method 2; (**c**)—Method 3; (**d**)—Method 4.

The two mentioned sets of derivatives, mentioned above, are spread out from their average value—the mean. A situation like this is possible if we did not find the extremum, or if the model function is not adequate, or the derivatives are huge in "any" small neighborhood of the extremum. In any case, the calculated values of variance are reasons to doubt the first two methods. Contrary to the third and fourth method the variances are not so huge numbers. As example we present toon Figure 6 the histogram of derivative values, computed for the first and fourth methods.



(**a**) (**b**)

Figure 6. Histogram and probability density function of normal distribution, generated for the set of all derivatives (computed in optimal values of parameters) with respect to y_i. (**a**): Method 1. (**b**): Method 4.

Based on the arguments above, one may consider Method 3 or Method 4. Preferably, Method 3 taking in account the mean absolute percentage error.

3.3. Verification of the Viscosity Prediction Ability of the Four Studied Methods

The 43 gas oils from Table 6 were selected in such a way to cover the whole possible diversity of properties of gas oils from primary and secondary origin which can encounter in any refinery all over the world. As was already mention in section "Results" Method 3 surpassed all other methods concerning the accuracy of viscosity prediction. Table 8 summarizes the statistical analyses for the four studied methods employing the data in Table 6. These data indubitably reveal the superiority of Method 3 as the best method to model gas oil viscosity. As a supplement the oil viscosity models of Aboul Seoud and Moharam [1] (Equation (6)), and Kotzakoulakis, and George [7] (Equation (65)), which are based on Walther's equation, were verified to predict viscosity of the 43 gas oils from Table 6. They predict the 43 gas oil viscosities with $\%AAD$ of 21.8%, and 89% respectively proving the superiority of Method 3 model.

$$lnln(VIS + 0.8) = 14.69ABP^{0.0684}SG^{0.267} - 3.682ln(T) \quad (65)$$

Table 8. Statistical analysis of the four studied methods and the models of Aboul Seoud and Moharam (Aboul Seoud and Moharam, 1999), and Kotzakoulakis and George (Kotzakoulakis and George, 2017) for the data from Table 6.

	Method 1	**Method 2**	**Method 3**	**Method 4**	**Aboul Seoud and Moharam**	**Kotzakoulakis and George**
Min E	−323.5	−389.2	−76.8	−112.8	−94.2	−729.9
Max E	17.6	19.6	20.5	28.1	35.2	57.2
RE	−2526.6	−2743.7	−480.1	−517.2	30.5	−291151
SE	2.6	2.7	1.8	2.2	2.7	7.1
RSE	28.3	29	19.9	23.6	28.9	77.3
SSE	44.1	57.6	3	5.7	3.7	141.1
%AAD	61.8	67.8	18.2	27.1	21.8	89
R2	0.9324	0.9323	0.9294	0.9281	0.9038	0.4352
Slope	0.771	0.7311	0.8669	0.7603	0.7209	0.8797
Intercept	3.66	3.93	1.48	1.75	1.5	3.45
AIC	192	159	9	153	204	316
BIC	201	168	18	162	215	326

The model obtained by Method 3 is currently used not only to predict viscosity of gas oils but also as a tool for verification of the correctness of viscosity measurement of gas oils in LUKOIL Neftohim Burgas Research laboratory. Several times it proved its usefulness as an indicator for incorrect viscosity measurement especially when H-Oil gas oils which

contain both high amount of aromatic compounds and relatively high content of waxes that makes problematic their viscosity measurement. Once HVGO viscosity at 80 °C was measured equal to 72 mm^2/s while the model based on Method 3 reported the value of 54 mm^2/s. The repetition of the viscosity measurement reported the value of 54 mm^2/s.

4. Conclusions

The gas oil properties average boiling point and specific gravity along with modified Walther's equation and nonlinear regression techniques can be used to model oil physical property viscosity. The four nonlinear regression techniques: least squares of absolute errors, least absolute errors, least squares of relative errors, and least absolute relative errors can model gas oil viscosity. The developed gas oil viscosity models by use of the four nonlinear regression methods showed comparable accuracy of viscosity calculation for the initial base of 41 vacuum gas oils. The statistical parameters relative error, standard error, relative standard error, sum of square errors, % average absolute deviation, coefficient of determination were not in position to unequivocally select the best model. Both *AIC*, *BIC* and the standard deviations of derivatives unambiguously indicated that the model developed by nonlinear regression least squares of relative errors was the best one. The sensitivity analysis with respect to given data also revealed that the LSRE model is the most stable one with the lowest values of standard deviations of derivatives.

The LSRE model demonstrated the highest accuracy of viscosity prediction of 43 gas oils not included in the initial data base. It was also superior in oil viscosity prediction relative to other published models based on modified Walther's equation. The LSRE can be used not only to predict gas oil viscosity but also to examine the correctness of the oil viscosity measurement.

Author Contributions: Conceptualization, E.S.; Data curation, S.N. and R.D.; Formal analysis, D.Y.; Investigation, D.D.S. (Denis D. Stratiev) and L.T.-Y.; Methodology, S.S., N.A.A.; Software, V.A., D.N., S.R. and D.D.S. (Danail D. Stratiev); Supervision, K.A.; Writing—original draft, D.S., S.N. and I.S. All authors have read and agreed to the published version of the manuscript.

Funding: This research was funded by Asen Zlatarov University–Burgas, Project: Information and Communication Technologies for a Digital Single Market in Science, Education and Security DCM # 577/17.08.2018 (2018–2021).

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: Not applicable.

Acknowledgments: The authors are grateful for the support provided by the Bulgarian Ministry of Education and Science under the National Research Programme "Information and Communi-cation Technologies for a Digital Single Market in Science, Education and Security" approved by DCM # 577/17 August 2018.

Conflicts of Interest: The authors declare no conflict of interest.

Nomenclature

ABP	Average boiling point
AIC	Akaike information criterion
ARI	Aromatic ring index

%AAD	% average absolute deviation
BIC	Bayesian information criterion
E	Error
FCC	Fluid catalytic cracking
HAGO	Heavy atmospheric gas oil
HCO	Heavy cycle oil
HTVGO	Hydrotreated vacuum gas oil
HVGO	Heavy vacuum gas oil
LAE	Least absolute errors
LARE	Least absolute relative errors
LCO	Light cycle oil
LSAE	Least squares of absolute errors
LSRE	Least squares of relative errors
LVGO	Light vacuum gas oil
MW	Molecular weight
NLLSR	Nonlinear least square regression
RE	Relative error
RI	Refractive index
RSE	Relative standard error
SA	Sensitivity analysis
SE	Standard error
SG	Specific gravity
SLO	Slurry oil
SRHVGO	Straight run heavy vacuum gas oil
SRLVGO	Straight run light vacuum gas oil
SRVGO	Straight run vacuum gas oil
SSE	Sum of square errors
VBGO	Visbreaker gas oil
VGO	Vacuum gas oil
υ	Kinematic viscosity, mm^2/s

References

1. Aboul-Seoud, A.L.; Moharam, H.M. A generalized viscosity correlation for undefined petroleum fractions. *Chem. Eng. J.* **1999**, *72*, 253–256. [CrossRef]
2. Abutaqiya, M.I.L.; Alhammadi, A.A.; Sisco, C.J.; Vargas, F.M. Aromatic Ring Index (ARI): A characterization factor for nonpolar hydrocarbons from molecular weight and refractive index. *Energy Fuels* **2021**, *35*, 1113–1119. [CrossRef]
3. Hernández, E.A.; Sánchez-Reyna, G.; Ancheyta, J. Comparison of mixing rules based on binary interaction parameters for calculating viscosity of crude oil blends. *Fuel* **2019**, *249*, 198–205. [CrossRef]
4. Hosseinifar, P.; Jamshidi, S. A new correlative model for viscosity estimation of pure components, bitumens, size-asymmetric mixtures and reservoir fluids. *J. Petrol. Sci. Eng.* **2016**, *147*, 624–635. [CrossRef]
5. Kamel, A.; Alomair, O.; Elsharkawy, A. Measurements and predictions of Middle Eastern heavy crude oil viscosity using compositional data. *J. Petrol. Sci. Eng.* **2019**, *173*, 990–1004. [CrossRef]
6. Kariznovi, M.; Nourozieh, H.; Abedi, J. Measurement and modeling of density and viscosity for mixtures of Athabasca bitumen and heavy n-alkane. *Fuel* **2013**, *112*, 83–95. [CrossRef]
7. Kotzakoulakis, K.; George, S.C. A simple and flexible correlation for predicting the viscosity of crude oils. *J. Pet. Sci. Eng.* **2017**, *158*, 416–423. [CrossRef]
8. Kumar, R.; Maheshwari, S.; Voolapalli, R.K.; Upadhyayul, S. Investigation of physical parameters of crude oils and their impact on kinematic viscosity of vacuum residue and heavy product blends for crude oil selection. *J. Taiwan Inst. Chem. Eng.* **2021**, *120*, 33–42. [CrossRef]
9. Malta, J.A.M.S.C.; Calabrese, C.; Nguyen, T.B.; Trusler, J.P.M.; Vesovic, V. Measurements and modelling of the viscosity of six synthetic crude oil mixtures. *Fluid Phase Equilibria* **2020**, *505*, 112343. [CrossRef]
10. Mehrotra, A.K. A Simple Equation for Predicting the Viscosity of Crude-Oil Fractions. *Chem. Eng. Res. Des.* **1995**, *73*, 87–90.
11. Pabón, R.E.C.; Filho, C.R.S. Crude oil spectral signatures and empirical models to derive API gravity. *Fuel* **2019**, *237*, 1119–1131. [CrossRef]
12. Raut, B.; Patil, S.L.; Dandekar, A.Y.; Fisk, R.; Maclean, B.; Hice, V. Comparative study of compositional viscosity prediction models for medium-heavy oils. *Int. J. Oil Gas Coal Technol.* **2008**, *1*, 229. [CrossRef]
13. Sánchez-Minero, F.; Sánchez-Reyna, G.; Ancheyta, J.; Marroquin, G. Comparison of correlations based on API gravity for predicting viscosity of crude oils. *Fuel* **2014**, *138*, 193–199. [CrossRef]

14. Stratiev, D.S.; Nenov, S.; Shishkova, I.K.; Dinkov, R.K.; Zlatanov, K.; Yordanov, D.; Sotirov, S.; Sotirova, E.; Atanassova, V.; Atanassov, K.; et al. Comparison of Empirical Models to Predict Viscosity of Secondary Vacuum Gas Oils. *Resources* **2021**, *10*, 82. [CrossRef]
15. Samano, V.; Tirado, A.; F´elix, G.; Ancheyta, J. Revisiting the importance of appropriate parameter estimation based on sensitivity analysis for developing kinetic model. *Fuel* **2020**, *267*, 117113. [CrossRef]
16. Ghorbani, B.; Hamedi, M.; Shirmohammadi, R.; Mehrpooy, M.; Hamedi, M.H. A novel multi-hybrid model for estimating optimal viscosity correlations of Iranian crude oil. *J. Petrol. Sci. Eng.* **2016**, *142*, 68–76. [CrossRef]
17. Khamehchi, E.; Mahdiani, M.R.; Amooie, M.A.A.; Hemmati-Sarapardeh, A. Modeling viscosity of light and intermediate dead oil systems using advanced computational frameworks and artificial neural networks. *J. Petrol. Sci. Eng.* **2020**, *193*, 107388. [CrossRef]
18. Saltelli, A.; Tarantola, S.; Campolongo, F.; Ratto, M. *Sensitivity Analysis in Practice*; John Wiley & Sons Ltd.: Chichester, UK, 2004.
19. Parhamifar, E.; Tyllgren, P. Assessment of asphalt binder viscosities with a new approach. In Proceedings of the E&E Congress 2016 | 6th Eurasphalt & Eurobitume Congress, Prague, Czech Republic, 1–3 June 2016.
20. Castillo, E.; Hadi, A.S.; Conejo, A.; Fernndez-Canteli, A. A general method for local sensitivity analysis with application to regression models and other optimization problems. *Technometrics* **2004**, *46*, 430–444. [CrossRef]
21. Alcazar, L.A.; Ancheyta, J. Sensitivity analysis based methodology to estimate the best set of parameters for heterogeneous kinetic models. *Chem. Eng. J.* **2007**, *128*, 85–93. [CrossRef]
22. Diarov, I.N.; Batueva, I.U.; Sadikov, A.N.; Colodova, N.L. *Chemistry of Crude Oil*; Chimia Publishers: St.Peterburg, Russia, 1990; Volume 51. (In Russian)
23. Fisher, I.P. Effect of feedstock variability on catalytic cracking yields. *Appl. Catal.* **1990**, *65*, 189–210. [CrossRef]
24. Walther, C. Ueber die Auswertung von Viskosit€atsangaben. *Erdoel Teer* **1931**, *7*, 382–384.
25. Solodov, M.V.; Svaiter, B.F. A globally convergent inexact Newton method for systems of monotone equations. In *Reformulation: Nonsmooth, Piecewise Smooth, Semismooth and Smoothing Methods*; Springer: Boston, MA, USA, 1998; pp. 355–369.
26. Zhou, W.J.; Li, D.H. A globally convergent BFGS method for nonlinear monotone equations without any merit functions. *Math. Comput.* **2008**, *77*, 2231–2240. [CrossRef]
27. Demidenko, E. Is This the Least Squares Estimate? *Biometrika* **2000**, *87*, 437–452. [CrossRef]
28. Takayama, A. *Mathematical Economics*; Cambridge University Press: New York, NY, USA, 1985; ISBN 0-521-31498-4.
29. Burnham, K.P.; Anderson, D.R. *Model Selection and Multimodel Inference*, 2nd ed.; Springer: New York, NY, USA, 2002.