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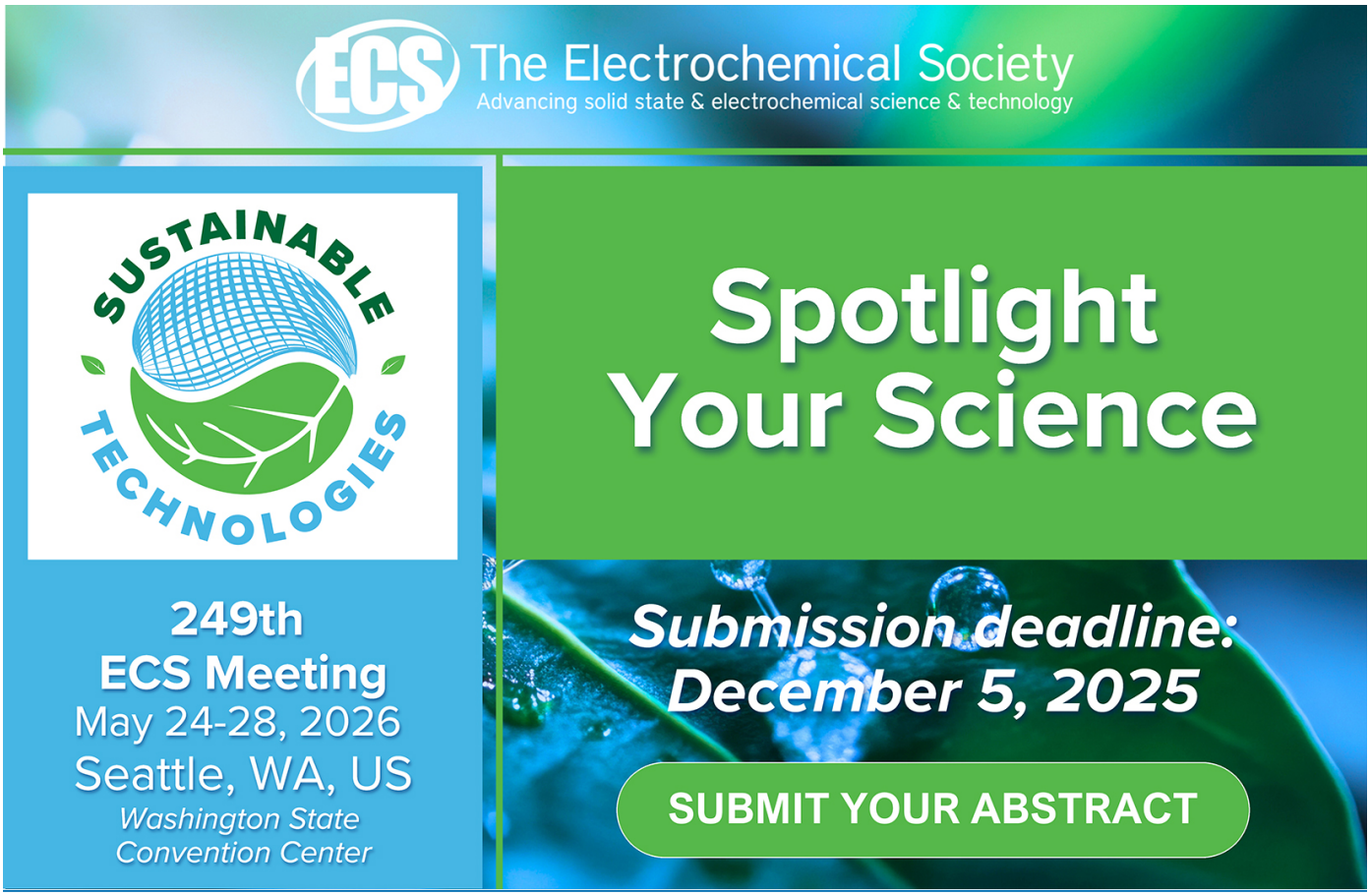

Semi-empirical correlations and an artificial neural network for liquid dynamic viscosity of low GWP refrigerants

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Abstract. In this work, simple semi-empirical correlations to describe the temperature and the pressure dependence of the dynamic viscosity of low GWP refrigerants, namely HydroFluoroOlefins (HFOs) and HydroChloroFluoroOlefins (HCFOs), in the liquid phase are presented. Firstly, the experimental liquid dynamic viscosity data available in scientific literature and databases were collected and statistically analyzed. From the data collected for low pressures, the Latini et al. (2002, 1990) correlation for the dynamic viscosity of liquid refrigerants in saturated conditions was re-fitted and constants expressly dedicated to the studied low GWP refrigerants were obtained. Then, the proposed temperature-dependent correlation was modified to represent liquid dynamic viscosity dependence on pressure. In addition, an artificial neural network was developed to predict the dependence of the liquid viscosity of the studied refrigerants on temperature and pressure. This model was trained, validated, and tested for the selected dataset. The results of the proposed correlations and the multi-layer perceptron neural network were compared with the liquid viscosity calculations provided by some of the most well-known literature correlations and REFPROP 10.0, proving the accuracy of the proposed models for engineering applications.

1. Introduction

In recent years, a worldwide search is underway to find low global warming potential (GWP) replacements, also known as “fourth generation” refrigerants [1], to conventional refrigerants which are long-lived greenhouse gases (GHGs). The aim of this search is to achieve GHG emission limitation targets set by environmental constraints and regulations [2,3]. Currently, only a limited number of low GWP synthetic refrigerants, mostly HydroFluoroOlefins (HFOs) and HydroChloroFluoroOlefins (HCFOs), have been identified as having suitable safety, environmental, and thermodynamic characteristics for different refrigeration applications [1]. However, most of the selected alternatives require a trade-off between flammability and suitable thermodynamic characteristics. In addition, a limited number of experimental data for the thermophysical properties of the potential low GWP refrigerants, such as saturated pressures, *pvT*, dynamic viscosity, and surface tension, are available in the literature [4]. Therefore, it is necessary to develop models that estimate their properties with a sufficient level of accuracy.

The viscosity of fluids is used in the design and in quality control of several scientific and industrial processes and operations. In particular, the viscosity of refrigerants is one of the transport properties required for detailed analyses of system components for refrigeration systems. Because of the complex nature of the viscosity of liquids, there is no comprehensive theoretical basis able to accurately calculate

this property. To overcome the shortcoming, various empirical and semi-empirical correlations and estimation methods for the liquid dynamic viscosity have been developed and have been presented in the literature [5,6]. Many of these models have been developed to estimate the dependence of liquid viscosity on temperature; instead, few models are also able to accurately represent its pressure dependence.

For refrigerants and their mixtures, different models have been developed to represent their viscosity behavior in the liquid phase. Some of the main models available in the literature are as follows: fluid-specific correlations and extended corresponding states (ECS) methods used in REFPROP 10.0 [7]; models based on the residual entropy scaling approach [8,9]; simple semi-empirical estimation methods for the dynamic viscosity of liquid refrigerants and their mixtures in saturated conditions developed by Latini and his co-workers [10,11]; a viscosity model based on Patel-Teja equation of state [12]; a semi-empirical model for the viscosity of liquid refrigerants and their mixtures based on the Modified Enskog Theory [13]; feed-forward artificial neural network (ANN) models [14].

This work presents modified versions of the simple correlations presented by Latini and his co-workers to describe the dependence of the dynamic viscosity of low GWP refrigerants on temperature and pressure. The modified correlations were developed using the experimental liquid viscosity data collected from the scientific literature and databases. Moreover, a feed-forward ANN model, known as multi-layer perceptron neural network, is presented to describe the temperature and pressure dependence of the liquid dynamic viscosity for the studied alternative refrigerants. Finally, the results provided by the proposed models are compared with that given by some models available in the literature.

2. Data selection and analysis

A total of 794 experimental liquid dynamic viscosity (η) data for 7 potential low GWP halogenated alkene refrigerants, namely R1233zd(E), R1234yf, R1234ze(E), R1234ze(Z), R1224yd(Z), R1336mzz(Z) and R1336mzz(Z), were selected through a literature survey. The η data were selected by the following fluid by fluid analysis and critical selection: the values showing deviations more than three sigma from the mean values were rejected, together with the data clearly beyond the common trend and the data without details about measurement conditions (i.e., pressures and densities). In addition, the experimental data measured at $T_r > 0.9$ ($T_r = T\,T_c^{-1}$, where T_c is the critical temperature) were neglected since, in general, simple estimation models are not able to provide accurate descriptions of the viscosity behavior of liquids near the critical point. Table 1 summarizes the number of selected data for each fluid and their temperature, pressure, and dynamic viscosity ranges, together with the data source references.

Table 1. Summary of the selected experimental liquid dynamic viscosity data for the studied low GWP refrigerants.

Refrigerant	Selected n. p.	T range, K	p range, MPa	η range, mPa s	Source
R1233zd(E)	155	243.14 – 393.57	0.15 – 40.0	0.112 – 0.732	[15–18]
R1234yf	154	243.17 – 329.98	0.10 – 30.0	0.109 – 0.393	[19–23]
R1234ze(E)	149	243.17 – 343.16	0.06 – 30.0	0.109 – 0.517	[20–22,24]
R1234ze(Z)	49	312.45 – 374.12	0.50 – 4.07	0.110 – 0.224	[25]
R1224yd(Z)	76	303.06 – 384.92	1.00 – 4.05	0.106 – 0.296	[26,27]
R1336mzz(E)	33	302.20 – 353.43	0.23 – 4.02	0.143 – 0.271	[28,29]
R1336mzz(Z)	178	253.14 – 394.03	0.09 – 40.0	0.128 – 0.958	[28,30,31]

Figure 1 shows the behaviors of the 794 selected points for the 7 analyzed refrigerants as a function of reduced temperature (T_r) and reduced pressure (p_r). Figure 1 clearly depicts that the liquid viscosity of the selected fluids increases with reduced pressure. However, as shown by the filled symbols in Figure 1, the pressure has a very limited effect on η up to moderate pressures and the η values up to $p_r = 1$ are almost constant at a fixed T_r.

Almost all the refrigerants' physical properties used in this work were collected from REFPROP 10.0 [7]. The physical properties of the following fluids were selected from other sources: R1224yd(Z) [32], R1336mzz(E) [33] and R1336mzz(Z) [34].

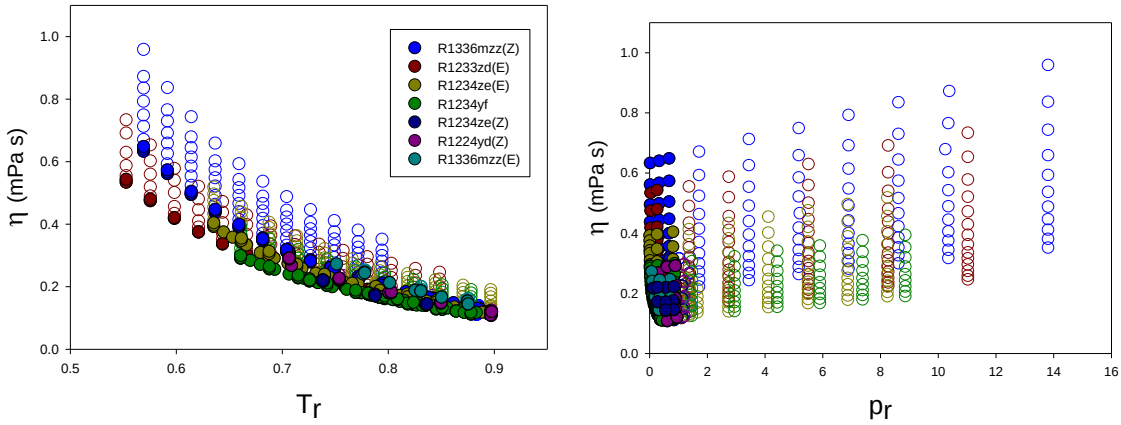

Figure 1. Behaviours of the selected liquid dynamic viscosity data for halogenated alkene refrigerants as a function of reduced temperature (T_r) (left) and reduced pressure (p_r) (right). The filled symbols represent the points measured at $p_r < 1$ and the open symbols represent the points measured at $p_r > 1$.

3. Estimation of the liquid dynamic viscosity

This section presents literature estimation methods for the liquid dynamic viscosity of wide families of fluids, together with modifications of some of them to describe the temperature dependence and pressure dependence of liquid dynamic viscosity of the studied alternative refrigerants. Finally, the architecture and characteristics of the proposed ANN model are described.

3.1. Literature estimation methods

The literature estimation methods suitable for wide families of fluids, especially refrigerants, and characterized by easily available physical properties were analyzed. Therefore, the regressions having fluid-specific constants obtained from the experimental data to describe the viscosity of each liquid as a function of the temperature were not considered. Similarly, many literature estimation methods requiring unavailable input properties for the studied refrigerants [5,6] were not analyzed. Consequently, only few estimation methods showing a reasonable compromise between simplicity and generality were considered. A brief description of some of these models is provided below.

Dutt [35] developed a general correlation to estimate the kinematic viscosity (η/ρ) of petroleum crude oil fraction defined as:

$$\ln\frac{\eta}{\rho} = -3.0171 + \frac{442.78 + 1.6452T_B}{T + (239 - 0.19T_B)} \qquad (1)$$

where η is the liquid dynamic viscosity in mPa s, ρ is the density in g mL^{-1}, T is the temperature in °C, and T_B is the normal boiling point in °C.

Latini and his co-workers [10,11] presented semi-empirical correlations to describe the temperature dependence of the liquid dynamic viscosity of different families of refrigerants and their mixture at the saturated conditions. The authors developed the correlations by combining the Batschinski equation [36] modified by Hildebrand [37] with the rule of Mathias [38] that links the reduced temperature and the density at the normal boiling point with the sum of liquid and vapour densities. The following expression is the most recent form of the correlations proposed by Latini:

$$\frac{1}{\eta} = A\left(\frac{1}{C - T_r} - 1\right) \qquad (2)$$

where A and C are constants. While C is regressed from the experimental data, the authors defined the constant A as:

$$A = hM^{\alpha}T_{Br}^{\beta} \qquad (3)$$

where M is the molecular mass, $T_{Br} = T_B\ T_C^{-1}$ is the reduced normal boiling point temperature, and h, α, β are regressed constants. The constants of Equation (2) and Equation (3) were originally calculated for both the methane and ethane series of refrigerants and ensured average absolute deviations of the liquid dynamic viscosity (AARD(η)) generally less than 6%, where AARD (η) is defined as:

$$AARD(\eta) = \frac{100}{N}\sum_{i=1}^{N}\frac{\left|\eta_{\exp,i} - \eta_{calc,i}\right|}{\eta_{\exp,i}} \qquad (4)$$

Recently, a semi-empirical model for the liquid dyamic viscosity of various refrigerants, among which different HFOs and HCFOs, and their mixtures was developed by Liu et al. [13] based on the Modified Enskog Theory. It has the following form:

$$\eta = \eta_0 b'\rho\left(\frac{1}{b'\rho g(\sigma)} + B + 0.761 b'\rho g(\sigma)\right) \qquad (5)$$

where η_0 is the dilute gas viscosity, ρ is the molar density, b' is the covolume, $g(\sigma)$ is the radial distribution function, and B is a temperature and pressure-dependent empirical parameter specific for each fluid. As in the original work, η_0 was calculated by the Chapman-Enskog solution of the Boltzmann equation assuming that interactions between molecules can be roughly simulated by those of Lennard-Jones particles, while b' and $g(\sigma)$ were calculated from the Peng-Robinson equation of state [39]. Moreover, the expression and coefficients of B presented in the original work were used here. As pointed out by the authors, the presented model showed high accuracy and simplicity for the analyzed refrigerants when compared with other literature models.

3.2. Modified versions of the Latini correlation

To provide a better η representation for the studied liquid refrigerants, a modified version of Equation (2) developed by Latini and his co-workers is proposed. In particular, the constants of Equation (2) and Equation (3) were regressed for the alternative refrigerants by minimizing the root mean square errors of the liquid dynamic viscosity (RMSE(η)) between the selected experimental data and the calculated values. Since the model is only suitable for liquids at saturated conditions, the constants were regressed on the experimental η measured at reduced pressure up to 1 (a total of 407 data points). The value obtained are reported in Table 2.

Table 2. Constants proposed in this work for Equation (2) and Equation (5).

Equation	C	h	α	β	D	E
(2)	1.376	1.387	-0.05275	-4.9	-	-
(6)	1.376	1.387	-0.05275	-4.9	0.9758	1.139

To describe the complete experimental dataset of the studied low GWP refrigerants (794 data points), Equation (2) was further modified to take into account the pressure dependence of η. Considering that only few estimation methods representing the liquid dynamic viscosity dependence on pressure have been proposed in the literature [5,6] we analyzed different models to find the most accurate results. The following modified version of the Latini correlation based on the Lucas model for the effect of the pressure [40] gave the lowest deviations for the selected dataset:

$$\frac{1}{\eta} = A\left(\frac{1}{C - T_r} - 1\right)\frac{1 + D\cdot p_r}{1 + p_r^E} \qquad (6)$$

where D and E are regressed constants and A is defined as in Equation (3). Thus, Equation (6) is the product of the saturation phase contribution for a pressure-dependent factor. This factor has a similar form to the Lucas model; however, its expression is simpler than that of the original equation since the reduced temperature and the acentric factor are not included. C, h, α, β were kept equal to the values obtained for dataset up to $p_r < 1$. The values of D and E were obtained by minimizing the RMSE(η) for the complete dataset (794 data points) and their values are reported in Table 2.

3.3. Artificial neural network

Neural networks are one of the most important approaches that can be used for the prediction of a thermophysical property. In general, neural networks are a mathematical construction based on human brain processing. They are trained to recognize certain patterns until the data given as input are matched to the outputs. In several literature works, neural networks were used for surface tension [41] or thermal conductivity [42] and different applications for viscosity [14].

Different types of networks can be used, such as perceptrons, radial basis, or neural networks with linear models or feed-forward networks. In our case, we used a feed-forward network with only one layer of neurons. During the training of the proposed neural network, the initial dataset was divided into the following three parts to avoid overfitting problems and evaluate its prediction capability: training, validation, and test datasets. The first dataset was used in the internal training process to calculate the hyperparameters of the network. The validation dataset was used by the network to validate the internal training process. Finally, the test dataset was used to test the network's capability to predict new data. In fact, this corresponds to a part of the dataset that the network does not see during the training process. As can be seen from Figure 2, the differences decrease as the number of neurons increases until they reach AARD(η) = 0.79% of the 23rd neuron, which is the one chosen for the architecture of the neural network.

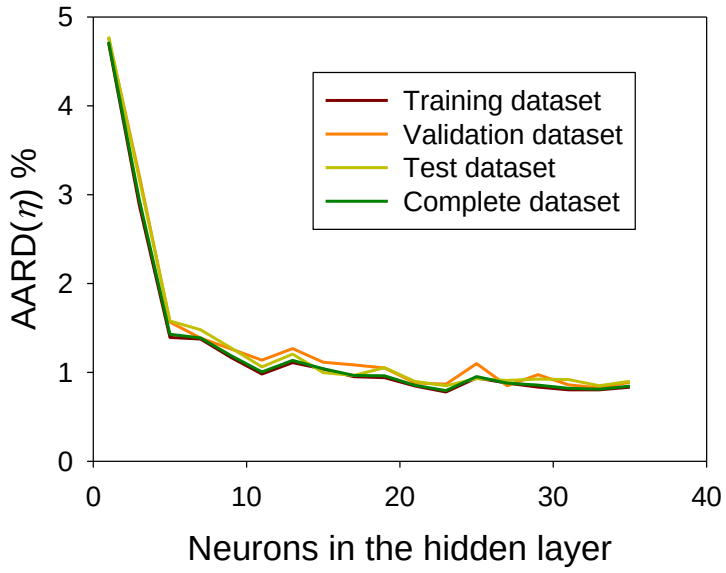

Figure 2. AARD(η) values for the complete dataset, for the training, validation, and test datasets, as a function of the number of neurons.

4. Results and discussion

Equation (2) with the regressed constants of Table 2 ensured an accurate description of the liquid dynamic viscosity for the studied low GWP refrigerants at reduced pressure up to 1 (407 data points),

giving AARD(η) equal to 3.96 % and maximum absolute relative deviations (MARD(η)) equal to 15.13 %.

The results provided by Equation (6) with the regressed constants of Table 2 are reported in Table 3 and in Figure 3.

These results show that the modified version of the Latini correlation is able to describe the pressure dependence of the dynamic viscosity for the studied liquid refrigerants. In fact, despite it provided deviations higher than 20 % for some points, most of the deviations between the experimental and calculated values are within 10 %. It is worth noting that Equation (6) (AARD (η) = 3.76%) continues to provide comparable results to those of Equation (2) (AARD (η) = 3.96%) for the experimental values at p_r <1 (407 data points) while taking pressure into account. Therefore, Equation (6) constitutes a good approximation of the whole range of pressures studied.

As we can see from Table 3, the results of the neural network are very satisfactory considering that the neural network presented is relatively simple. In fact, the AARD(η) is equal to 0.79% for the complete dataset and fluids with many points are also well represented. As concern the MARD(η), it can be clearly seen that it never exceeds 7.39 %. This shows that the network is very good at predicting the viscosity of HFO refrigerants.

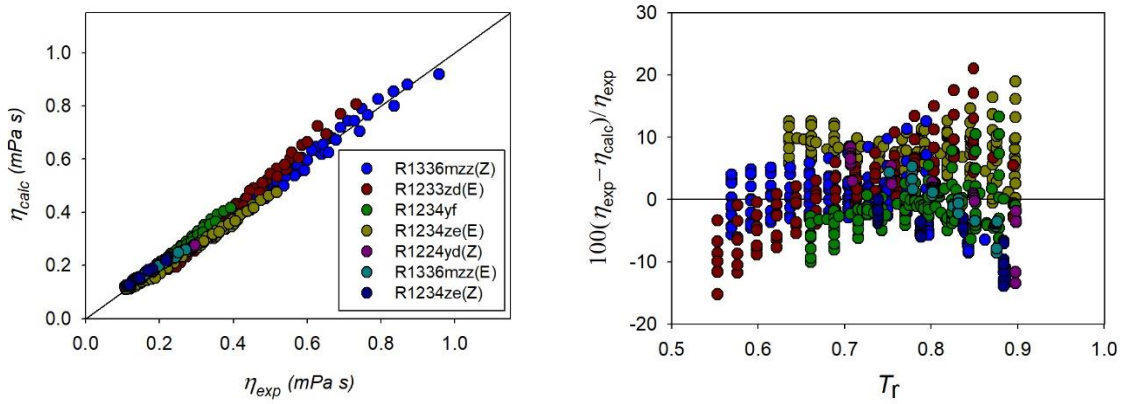

Figure 3. Liquid dynamic viscosities calculated with Equation (6) versus the selected experimental liquid viscosity data (left) and deviations between the complete experimental dataset and calculated values provided by Equation (6) versus the reduced temperature (right).

Finally, the selected liquid dynamic viscosity data for the halogenated alkene refrigerants were compared with the values provided by Equation (1), Equation (5) and REFPROP 10.0. The density values used in Equation (1) and Equation (5) for the analyzed fluids were calculated from REFPROP 10.0. It was not possible to perform the comparison between the data of R1336mzz(E) and the calculations of the selected models since this fluid is not available in REFPROP 10.0 database yet.

Table 3. Average absolute relative deviations (AARD(η)%) and maximum absolute relative deviations (MARD(η)%) between the selected experimental data and the values provided by Equation (1), Equation (5), Equation (6), artificial neural network (ANN) and REFPROP 10.0.

Refrigerant	Selected n. p.	Eq. (1) AARD(η)% MARD(η)%	Eq. (5) AARD(η)% MARD(η)%	Eq. (6) AARD(η)% MARD(η)%	ANN AARD(η)% MARD(η)%	REFPROP 10.0 AARD(η)% MARD(η)%
R1233zd(E)	155	29.09 63.95	8.42 17.08	4.42 21.07	0.65 7.39	1.63 9.22
R1234yf	154	38.92 67.53	2.06 8.96	2.91 13.29	0.96 4.92	1.35 6.85
R1234ze(E)	149	27.47 65.95	1.72 4.60	7.85 18.95	1.14 5.12	1.87 6.69

R1234ze(Z)	49	51.81 73.23	1.83 6.07	5.20 13.93	0.88 3.44	1.27 4.01
R1224yd(Z)	76	52.24 88.92	1.79 7.24	3.39 13.38	0.95 3.80	1.44 4.37
R1336mzz(E)	33	- -	- -	2.90 8.51	0.73 2.42	- -
R1336mzz(Z)	178	25.45 67.53	2.28 8.22	3.38 13.45	0.41 2.79	1.13 5.14
Overall	794	33.74 -	3.30 -	4.43 -	0.79 -	1.46 -

The obtained results are reported in Table 3 and Figure 4. As a general comment, it is evident that Equation (1) provided very high deviations and is not suitable for the description of the complete dataset. It has to be pointed out that this correlation was not oriented to refrigerants. Instead, accurate results were generally ensured by Equation (5) and REFPROP 10.0. This outcome was expected since REFPROP 10.0 calculates the dynamic viscosity using fluid-specific correlations or ECS methods for the studied refrigerants, while regressed coefficients specific for each refrigerant are used in Equation (5). However, higher deviations for R1233ze(E) were given by Equation (5). This could be due to the dataset analyzed here and the physical properties used in Equation (5) that are different from that used in the original work.

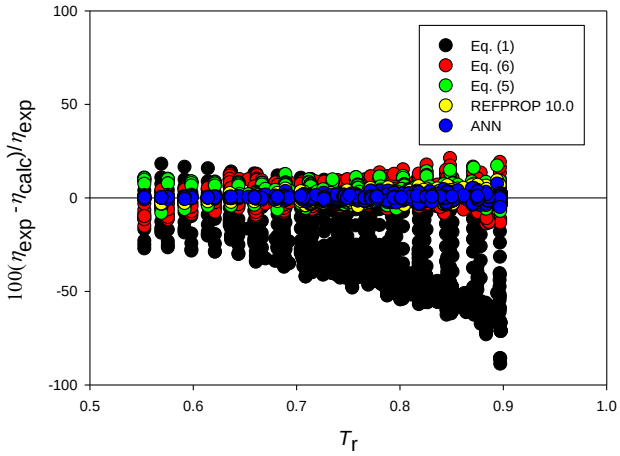

Figure 4. Deviations between the complete experimental dataset and values calculated from Equation (1), Equation (5), Equation (6), artificial neural network (ANN) and REFPROP 10.0 versus the reduced temperature.

5. Conclusions

This work presents a re-fitted and modified version of the correlation developed by Latini and his co-workers for the liquid viscosity description of the temperature dependence and pressure dependence of low GWP refrigerants. Despite few exceptions, the proposed correlation provides low deviations between the calculated and the selected experimental data, giving an AARD (η) = 3.76 % for reduced pressures up to 1 (407 data points) and an AARD (η) = 4.43 % for the complete dataset (794 data points). Therefore, it is reliable for the reduced temperatures up to 0.9 and the reduces pressures in the complete dataset of the refrigerants under investigation. In addition, an artificial neural network was developed to describe the temperature and pressure dependence of liquid viscosity for the studied fluids. As expected, very low deviations between the experimental data and the values calculated from the ANN were obtained (AARD (η) = 0.79 % for the complete dataset).

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