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Dynamic Viscosity of Partially Carbonated Aqueous Monoethanolamine (MEA) from (20 to 150) ℃

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ABSTRACT

Viscosities of liquid solutions of monoethanolamine (MEA), water (H_20), and carbon dioxide (CO_2) have been measured. The mass fraction of MEA was (10, 20, 30, 40, and 50)%, and CO_2 loading values was between (0.1 to 0.5) mol CO_2/mol MEA. The operating temperature was varied between (20 to 150) ℃. The available literature data for temperature range (25 to 80) ℃ for mass fraction of (20, 30 and 40)% were used to compare the measurement data. The dynamic viscosity increase with the increase of CO_2 loading and decrease with an increase of temperature. The measured data were compared with the data predicted from available regression equation for certain temperature range. Agreement between measured data and the correlation data was satisfactory.

Keywords: Monoethanolamine, Viscosity, CO_2 loading, Rheometer, Temperature effect

1. Introduction

The various types of amines such as Monoethanolamine (MEA), diethanolamine (DEA), N-Methyldiethanolomine (MDEA) have been used for gas treating systems in a wide variety of applications[1]. The information available in literatures related to the physical properties of amines is limited. The uses of physical properties are typically for calculating of column dimensions and mass transfer correlations in gas absorption process[2,3].

Moreover, dynamic viscosities of partially carbonated aqueous amines are rare to find in the literatures. However, some of the data are available with limited concentration values and limited temperature range. Weiland *et al.*[4] has performed the experiments for CO_2 loaded aqueous MEA for temperature for 25 ℃ with 10, 20, 30 and 40% mass concentration. However, they have performed the experiments only for temperature 25 ℃.

Scarcity of physical properties availability was motivated to perform the experiments. Solution viscosity is one of the main parameters when considering the gas absorption process. Most of the literatures are reported the data only to temperature 80 ℃ and for limited concentration values only. Amundsen *et al.*[5] has reported the viscosity data for temperature range from (25 to 80) ℃ for concentration of MEA 20, 30 and 40 % mass basis. However, they have considered the CO_2 loading values $\alpha \in$ [0.1, 0.5] for their experiments. Therefore, more viscosity values are missing in the range while considering the available data for aqueous MEA with CO_2 loaded. This reason motivated us to perform the experiments for this study.

Modeling and simulation of gas absorption process require number of parameter values that related to the mass transfer rate[4]. This work was taken to determine the dynamic viscosity of Monoethanolamine (MEA) which is mainly considered for gas absorption. The CO_2 loaded amine viscosities are analyzed for the solutions with the mass ration, $r \in$ [0.1, 0.5] and CO_2 loading, $\alpha \in$ [0.1, 0.5] at temperatures, $T \in$ [20, 150] ℃. The measured data were compared with the available literature data[5]. At the same time, data were compared with the correlation presented in Weiland *et al.*[4].

2. Experimental section

The pure MEA was purchased from Merck KGaA supplier. The purity of the amine is 99.5% (mass basis). Amine solutions are prepared to get total concentration of amine mixture as 10%, 20%, 30%, 40% and 50% mass basis. The mixture after adding amines and distilled water is well stirred to get uniformity of the solution.

Aqueous solutions of amines were prepared using degassed, purified water and amines. Sample concentration maintained by adding required portion of amine and water with the help of analytical balance that has an accuracy of $\pm 1 \cdot 10^{-7}$.The high CO_2 loaded samples, α = (>0.5), prepared by bubbling CO_2 gas through an unloaded solution at required mass flow rate of CO_2 for an appropriate period. The required CO_2 loaded samples were prepared by diluting

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of high CO_2 loading with an unloaded solution in an appropriate ratio to get required loading values, α = (0.1 to 0.5).

The high loaded amine solutions were analyzed using titration method to check the exact CO_2 loading value and the amine concentration. The 1 mol $Ł^{-1}$ HCl solution is used to perform the titration to check the mass concentration of the amine solution. The sample is prepared by adding 2 g of each prepared amine solution with de-ionized water until each sample became 100 cm^3 in total. The amount of amine present in the sample is calculated by using the amount of HCl used for the titration. The sample preparation for the loading analysis was carried out by mixing about (0.5 to 1.0) g of the loaded amine solution with 50 cm^3 each from 0.3 mol $Ł^{-1}$ $BaCl_2$ and 0.1 mol $Ł^{-1}$NaOH. Those samples were heated around 5 min to let CO_2 in the samples to react with $BaCl_2$ and make precipitate as $BaCO_3$, then cooled down in a water bath. Moreover, the precipitate is collected and added to the 100 cm^3 of de-ionized water and then titrated with 0.1 mol $Ł^{-1}$HCl solution until the mixture reached the equilibrium point. The mixture was heated to remove the all of the dissolved CO_2. Then, same mixture was used for back titration with 0.1 mol $Ł^{-1}$NaOH solution to check the amount of excess HCl. At last, the moles of HCl reacted with $BaCO_3$ precipitate was used to find the amount of CO_2 in the corresponding partially carbonated aqueous amine sample and subsequently the CO_2 loading value of the sample.

Dynamic viscosity was measured using MCR 101 Anton Paar double-gap rheometer. The viscometer was calibrated against the petroleum distillate and mineral oil calibration fluid which is purchased from Paragon Scientific Ltd. The calibration factor was decided according to the experimental value and given literature value. The low-temperature measurements (20 – 30) ℃ were achieved by applying cooling system Physica VT2 connected together with the rheometer setup. Without further purification, all these amines were used for experimental studies. The temperature range from (40 – 150) ℃ is measured without cooling system. Two different pressure values are used for the measuring process in order to avoid the water vaporization at high temperatures. First part of the process (20 – 80) ℃ was completed with pressure 1.01 bar and the second part of the process (90 – 150) ℃ is completed with a 4.5 bar pressure.

The measured data for the different amines are compared with those available from the literature. The results that are obtained from the experiments were compared with the regression viscosity values which are predicted by correlations for viscosity at different temperatures.

3. Results and discussion

The viscosity data for partially carbonated MEA solutions, mass ratio r ∈ [0.1, 0.5], are presented in the **Table 1**-**Table 5** respectively. Five different concentration levels considered with five different CO_2 loading values for complete temperature range.

Temperature (℃)	**CO_2 loading (mol CO_2/mol MEA)**				
	α = 0.1	α = 0.2	α = 0.3	α = 0.4	α = 0.5
20	1.46	1.49	1.51	1.54	1.58
25	1.31	1.34	1.37	1.43	1.48
30	1.16	1.19	1.21	1.22	1.25
40	0.93	0.96	0.98	0.99	1.04
50	0.74	0.77	0.79	0.83	0.87
60	0.65	0.67	0.69	0.72	0.75
70	0.56	0.59	0.63	0.66	0.69
80	0.47	0.49	0.52	0.55	0.58
90	0.41	0.43	0.46	0.53	0.55
100	0.37	0.38	0.42	0.46	0.48
110	0.34	0.36	0.39	0.42	0.44
120	0.31	0.33	0.36	0.39	0.4
130	0.27	0.29	0.32	0.34	0.36
140	0.25	0.26	0.28	0.3	0.31

150	0.22	0.23	0.25	0.28	0.29

Table 1. Viscosity of MEA with different CO_2 loading, r = 0.1

Temperature (℃)	**CO_2 loading (mol CO_2/mol MEA)**				
	α = 0.1	α = 0.2	α = 0.3	α = 0.4	α = 0.5
20	2.09	2.15	2.34	2.46	2.68
25	1.81	1.86	1.94	2.16	2.27
30	1.58	1.62	1.68	1.88	1.98
40	1.27	1.34	1.42	1.48	1.63
50	1.02	1.08	1.17	1.21	1.28
60	0.85	0.89	0.99	1.02	1.08
70	0.72	0.75	0.82	0.85	0.90
80	0.61	0.65	0.70	0.72	0.75
90	0.52	0.6	0.68	0.7	0.72
100	0.47	0.57	0.64	0.66	0.69
110	0.41	0.51	0.56	0.61	0.64
120	0.37	0.48	0.5	0.54	0.58
130	0.33	0.44	0.48	0.52	0.54
140	0.3	0.37	0.42	0.46	0.48
150	0.27	0.33	0.37	0.39	0.42

Table 2. Viscosity of MEA with different CO_2 loading, r = 0.2

Temperature (℃)	**CO_2 loading (mol CO_2/mol MEA)**				
	α = 0.1	α = 0.2	α = 0.3	α = 0.4	α = 0.5
20	3.33	3.49	3.82	3.97	4.21
25	2.87	2.94	3.14	3.47	3.76
30	2.47	2.56	2.72	2.94	3.27
40	1.92	2.04	2.09	2.39	2.67
50	1.55	1.61	1.66	1.87	2.22
60	1.25	1.31	1.38	1.52	1.81
70	1.02	1.08	1.16	1.24	1.51

80	0.84	0.89	0.99	1.05	1.26
90	0.76	0.82	0.95	1.01	1.16
100	0.64	0.72	0.82	0.93	1.07
110	0.57	0.66	0.75	0.86	0.96
120	0.5	0.58	0.64	0.72	0.84
130	0.44	0.52	0.6	0.68	0.75
140	0.38	0.46	0.53	0.59	0.64
150	0.34	0.4	0.46	0.52	0.58

Table 3. Viscosity of MEA with different CO_2 loading, r = 0.3

Temperature (℃)	**CO_2 loading (mol CO_2/mol MEA)**				
	α = 0.1	α = 0.2	α = 0.3	α = 0.4	α = 0.5
20	5.01	5.56	6.70	7.49	7.89
25	4.25	4.82	5.39	6.16	6.98
30	3.56	4.10	4.61	5.22	5.84
40	2.74	3.02	3.28	3.63	4.05
50	2.13	2.37	2.65	2.97	3.22
60	1.64	1.82	2.15	2.52	2.72
70	1.30	1.52	1.77	2.05	2.37
80	1.07	1.24	1.39	1.71	1.94
90	1.01	1.16	1.24	1.66	1.84
100	0.93	1.05	1.11	1.42	1.65
110	0.84	0.96	0.99	1.21	1.43
120	0.73	0.84	0.86	0.96	1.1
130	0.67	0.74	0.79	0.85	0.98
140	0.56	0.61	0.68	0.76	0.84
150	0.45	0.53	0.58	0.6	0.66

Table 4. Viscosity of MEA with different CO_2 loading, r = 0.4

Temperature (℃)	CO_2 loading (mol CO_2/mol MEA)				
	α = 0.1	α = 0.2	α = 0.3	α = 0.4	α = 0.5
20	7.49	9.66	11.60	14.30	16.66
25	6.14	7.91	9.48	11.30	13.50
30	5.14	6.56	7.83	9.37	11.35
40	3.72	4.60	5.92	6.78	8.05
50	2.79	3.52	4.30	5.20	5.96
60	2.15	2.78	3.09	4.09	4.55
70	1.72	2.27	2.49	3.37	3.62
80	1.40	1.89	2.06	2.91	3.27
90	1.24	1.57	1.89	2.65	2.89
100	1.10	1.30	1.64	1.96	2.61
110	0.96	1.16	1.32	1.74	2.28
120	0.86	1.01	1.18	1.46	1.96
130	0.74	0.96	1.04	1.30	1.64
140	0.65	0.82	0.95	1.13	1.42
150	0.56	0.63	0.73	0.84	1.12

Table 5. Viscosity of MEA with different CO_2 loading, r = 0.5

The viscosity data measured for loaded mixtures at temperature from 25 to 80 ℃ are compared with data from Amundsen *et al*.[5] in **Figure 1** to **Figure 3** for three different amine concentration. The literature data only available for temperature up to 80 ℃ and concentration, r = (0.2, 0.3, 0.4).

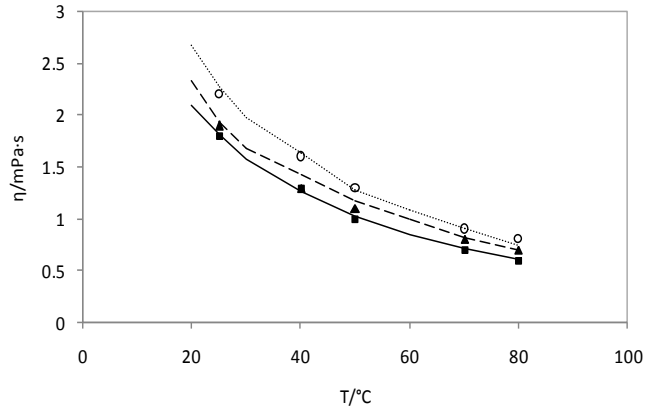

Figure 1. Viscosity variation with temperature, r = 0.2, Lines are experimental data: −, α = 0.1; - -, α = 0.3; ..., α = 0.5. Symbols refer to literature data: ■, α = 0.1; ▲, α = 0.3; ○, α = 0.5.

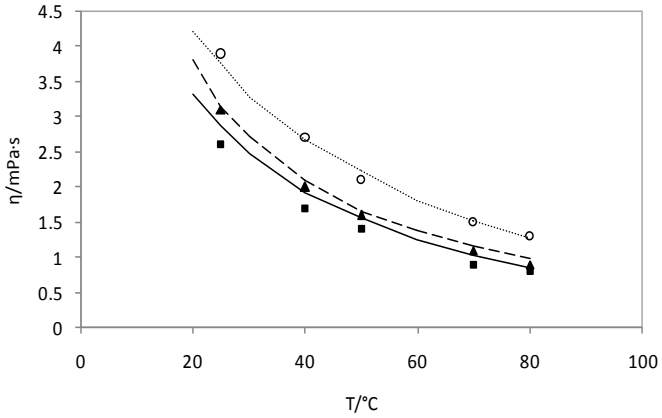

Figure 2. Viscosity variation with temperature, r = 0.3, Lines are experimental data: −, α = 0.1; - -, α = 0.3; ..., α = 0.5. Symbols refer to literature data: ■, α = 0.1; ▲, α = 0.3; ○, α = 0.5.

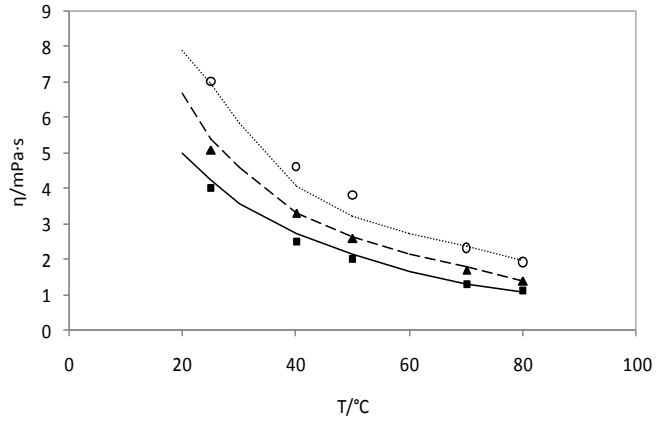

Figure 3. Viscosity variation with temperature, r = 0.4, Lines are experimental data: −, α = 0.1; - -, α = 0.3; ..., α = 0.5. Symbols refer to literature data: ■, α = 0.1; ▲, α = 0.3; ○, α = 0.5.

The literature data and experimental work in this study are in good agreement. The average absolute deviation (AAD) is 0.03 mPa s at r = 0.2, 0.04 mPa s at r = 0.3 and 0.003 mPa s at r = 0.4. The correlation suggested by Weiland *et al*. [4] is used to calculate the estimation values of viscosity for different temperatures (Equation 1).

$$\frac{\eta}{\eta_{H_2O}} = \exp\frac{\left[(aw+b)T+(cw+d)\right]\left[\alpha(ew+fT+g)+1\right]w}{T^2} \quad (1)$$

Where η and $\eta_{H2O}$are the viscosities of the amine mixture and water viscosity at that temperature respectively in mPa s. w is the mass percent of the solution, T is the operating temperature in K, and α is the CO_2 loading in amine mixture (mol CO_2/mol MEA). The required coefficients are given in the **Table 6**.

Parameter	Value for MEA
a	0
b	0
c	21.186
d	2373
e	0.01015
f	0.0093
g	-2.2589

Table 6. Parameters for solvent viscosity[4]

The equation can be used to calculate MEA solution viscosity up to amine concentration 40% mass basis with CO_2 loading up to 0.5 (mol CO_2/mol MEA) and to a maximum temperature 398 K[4]. Due to the limitations of applicability of the equation 1, experimental data are compared up to temperature 120 ℃ and r = (0.1, 0.2, 0.3, 0.4) solution concen-

tration. The required water viscosity for an above equation is taken from the previous studies[6]. **Figure 4, Figure 5**, **Figure 6** and **Figure 7** show the measurements from this work compared to the correlation viscosity values at T ∈ [20, 120] ℃ for amine solution mass ration, r ∈ [0.1, 0.4]. Figures show the viscosity (Y axis) variation with temperature (X axis) for different CO_2 loading values. The part of the experiential values is only compared with available regression viscosity values as it has limitations in equation.

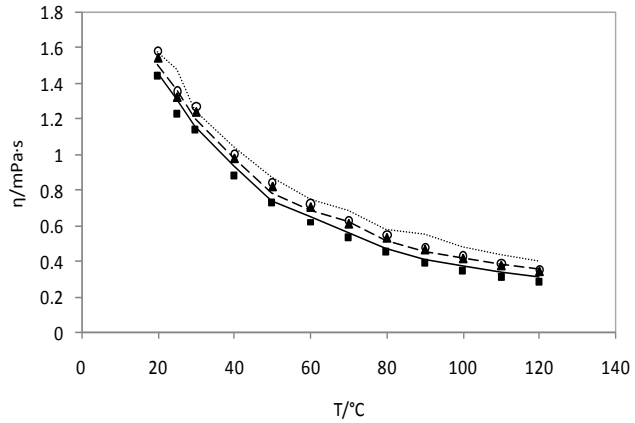

Figure 4. Viscosity variation with temperature, r = 0.1, Lines are experimental data: ─, α = 0.1; - -, α = 0.3; ..., α = 0.5. Symbols refer to regression data: ■, α = 0.1; ▲, α = 0.3; ○, α = 0.5.

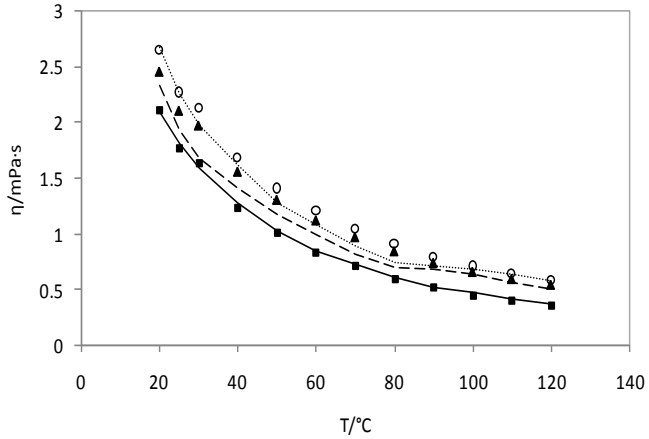

Figure 5. Viscosity variation with temperature, r = 0.2, Lines are experimental data: ─, α = 0.1; - -, α = 0.3; ..., α = 0.5. Symbols refer to regression data: ■, α = 0.1; ▲, α = 0.3; ○, α = 0.5.

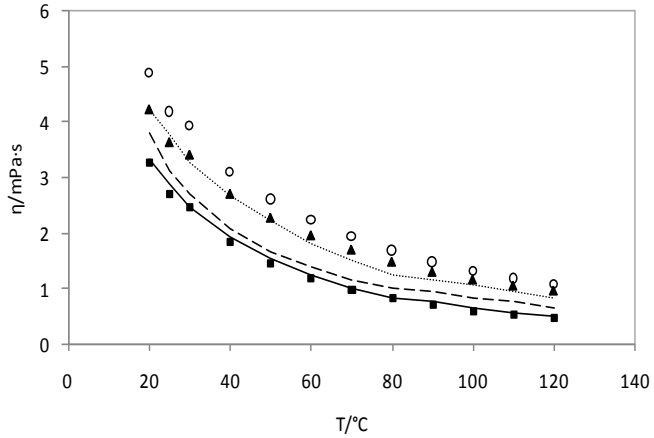

Figure 6. Viscosity variation with temperature, r = 0.3, Lines are experimental data: ─, α = 0.1; - -, α = 0.3; ..., α = 0.5. Symbols refer to regression data: ■, α = 0.1; ▲, α = 0.3; ○, α = 0.5.

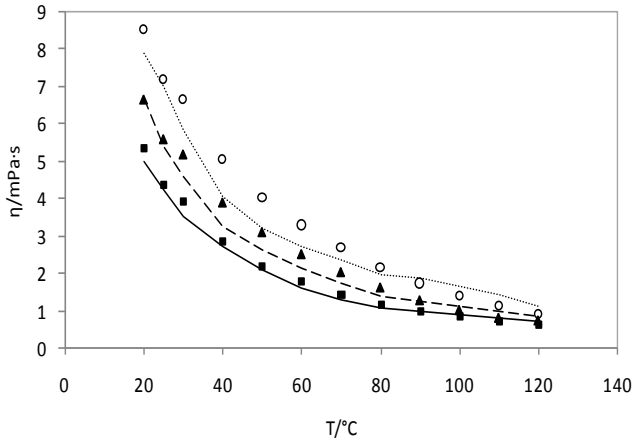

Figure 7. Viscosity variation with temperature, r = 0.4, Lines are experimental data: ─, α = 0.1; - -, α = 0.3; ..., α = 0.5. Symbols refer to regression data: ■, α = 0.1; ▲, α = 0.3; ○, α = 0.5.

The agreement between correlation results and this work in satisfactory. However, correlation shows over predicts for most of the viscosity values for every concentration. The average absolute deviation (AAD) between this work and equation regression data are 0.02, 0.09, 0.36, 0.19 mPa s respectively for mass fraction of amine, r = (0.1, 0.2, 0.3, 0.4).

4. Experimental uncertainties

The uncertainty of the viscosity measurements of CO_2 loaded aqueous amines arises as a combination of the uncertainty of the temperature measurements, sample preparation, CO_2 loading and measuring instrument uncertainties. The temperature accuracy, U(T), which is related to rheometer temperature controller, is given as ±0.3K. The maximum viscosity gradient against the temperature, $\Delta\eta/\Delta T$, is calculated as 0.040 mPa·s·K^{-1}. The corresponding uncertainty in η, $(\Delta\eta/\Delta T)\cdot\Delta T$, is then estimated as ±0.0120 mPa·s. The uncertainties of the sample preparation were found by calculating the error values (difference between the expected value and measured value r) of the prepared sample. The mass ratio uncertainty ±0.004, U(r), and the viscosity gradient ($\Delta\eta/\Delta r$) with 0.05 mPa·s are used for calculating the uncertainty of sample preparation. The resulting uncertainty in the sample preparation is calculated as, $(\Delta\eta/\Delta r).\Delta r$, ±0.00020. The uncertainty of loading, U(α), was found to be ±0.005 (mol CO_2/mol MEA) for MEA. The viscosity gradient, $\Delta\eta/\Delta\alpha$ was found as 2.1 mPa s (mol CO_2/ mol MEA)$^{-1}$. The corresponding uncertainty was calculated as $(\Delta\eta/\Delta\alpha)\cdot\Delta\alpha$, ±0.0105 mPa s. The rheometer accuracy is given as ± 0.002 mPa s. The overall uncertainty of η, U(η), is calculated by combining the partial uncertainties reported in this section with root sum of square method. The value is calculated as ±0.0161 mPa·s. The combined expanded uncertainty of the viscosity, Uc(η), is calculated as ±0.032 mPa s (level of confidence 0.95). The combined expanded uncertainty, suggested by symbol Uc, is obtained by multiplying overall uncertainty, U(η), by a coverage factor, suggested symbol k. Typically, k is assumed to be 2 with the level of confidence 0.95.

5. Conclusions

The dynamic viscosity of partially carbonated MEA solution was measured for the temperature range (20 to 150) ℃ for mass fraction (10 to 50)% and CO_2 loading (0.1 to 0.5) mol CO_2/mol MEA. The agreement with the literature data for temperature range (25 to 80) ℃ is satisfactory for mass fraction (20 to 40)%. The comparison between Weiland's proposed model and measurement data are in good agreement. However, regression model is valid only for mass fraction of MEA up to 40% and temperature up to 125 ℃. Therefore, measurement data were compared only for valid operating conditions. The AAD between this work and equation regression data are 0.02, 0.09, 0.36, 0.19 mPa s respectively for mass fraction of amine, r = (0.1, 0.2, 0.3, 0.4). However, Weiland's regression model can be used for estimation of viscosity inside the limitations. Further measurements have to perform for other amines as well.

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