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Densities, Excess Molar Volumes, Isothermal Compressibilities, and Isobaric Thermal Expansivities of the *N*-Methyldiethanolamine (1) + Water (2) System at Temperatures between (313 and 363) K and Pressures up to 20 MPa

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$P\rho T$ properties in the compressed liquid phase were measured for the system *N*-methyldiethanolamine (MDEA) (1) + water (2) at temperatures between (313 and 363) K and pressures up to 20 MPa. Densities of MDEA and four gravimetrically prepared mixtures of MDEA (1) + water (2), at $x_1 = 0.0369$, 0.0607, 0.0893, and 0.1302, were determined using a vibrating tube densimeter. The classical calibration method of the vibrating tube densimeter was used, using nitrogen and water as reference fluids. The uncertainty was estimated to be $\pm$ 0.2 kg·m^{-3} for the measured densities. The densities of each mixture and of MDEA were correlated using a volume explicit equation of six parameters. Isothermal compressibilities and isobaric thermal expansivities were calculated using the six-parameter equation with the correlated parameters obtained for MDEA and for the four mixtures. The uncertainties on these properties were estimated to be $\pm$ 0.005 GPa^{-1} and $\pm$ 5·10^{-7} K^{-1}, respectively. Excess molar volumes for the mixtures were determined using the measured densities of the mixture and MDEA volumes calculated from the six-parameter equation and water volumes calculated from a multiparameter reference equation of state (EoS). The uncertainty in excess molar volumes was estimated to be $\pm$ 0.006 cm^3·mol^{-1}.

Introduction

Alkanolamines and their mixtures with water have different applications as surfactants, detergents, personal care products, insecticides, cement, textiles, and agricultural products.[1,2] Also, alkanolamine aqueous solutions are important for the removal of acid gases, principally CO_2 and H_2S, from gas streams in natural gas processing, petroleum refining, and petrochemical manufacturing.[3,4] These washing processes are used in petroleum refining, coal gasification, and hydrogen production.[4] *N*-Methyldiethanolamine (MDEA) is a tertiary amine which was found to have the ability to separate selectively H_2S from CO_2.[5,6] Advantages of the use of MDEA solutions include their high loading capacity and low heat of reaction with acid gases, and MDEA does not degrade readily.[7] The lower heat of reaction leads to lower energy consumption.[7] For the design of suitable gas−liquid contactors of these processes, it is necessary that the density, viscosity, and other physical properties can be measured accurately.[8−10] These properties are useful in determining and interpreting other physicochemical properties such as liquid diffusivities, free-gas solubility, and the reaction kinetics of CO_2 with aqueous amine solutions.[11]

Concerning $P\rho T$ liquid data for the aqueous MDEA solutions at high pressures, only the work by Hawrylak et al.[12] reported volumetric properties of aqueous MDEA solutions over the temperature range of (283.15 to 524.57) K and pressures of (0.1, 10, and 20) MPa. Densities at atmospheric pressure have been measured more frequently for MDEA and for the MDEA + water system. Al-Ghawas et al.,[6] DiGuillo et al.,[2] Li and Shen,[8] Rinker et al.,[13] Li and Lie,[14] Welsh and Davis,[15] Wang et al.,[16] Hagewiesche et al.,[17] Hsu and Li,[18] Fischer et al.,[19] Henni et al.,[20] Aguila-Hernández et al.,[21] Maham et al.,[22] Bernal-Garcia et al.,[9] Weiland et al.,[11] Mandal et al.,[23] Rebolledo-Libreros and Trejo,[24] Paul and Mandal,[25] and Alvarez et al.[26] have measured densities at atmospheric pressure of MDEA and of the MDEA + water system.

The objective of this work was to measure the density of binary mixtures of MDEA (1) + water (2) at high pressure. Measurements were made over the concentration range of (0.20 to 0.50) mass fraction of MDEA, at temperatures from (313 to 363) K and pressures up to 20 MPa. Measurements reported in this work complement the data reported by Hawrylak et al.[12] The experimental density data were correlated with a six-parameter equation, and these correlations were used to calculate some derived thermodynamic properties. The excess molar volumes were calculated for the mixtures studied in this work.

Experimental Section

Materials. MDEA was obtained from Huntsman with a mass fraction purity of 0.99 containing a mass water fraction of 0.01. Water (HPLC grade) was obtained from Fisher with a stated mole fraction purity greater than 0.9995. Nitrogen (chromatographic grade) was from Air Products-Infra with a stated volume fraction purity of 0.99995. Nitrogen was used as received, and both liquids were used without further purification except for careful degassing under a vacuum with vigorous stirring.

Apparatus. The apparatus used in this work is based on that utilized by Galicia-Luna et al.[27] The configuration of the ancient apparatus was modified to determine VLE and $P\upsilon T$ properties

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Table 1. Experimental Liquid Densities, ρ, of MDEA

T/K = 313.09		T/K = 323.01		T/K = 332.95		T/K = 342.78		T/K = 352.68		T/K = 362.52	
p / MPa	ρ / kg·m^{-3}	p / MPa	ρ / kg·m^{-3}	p / MPa	ρ / kg·m^{-3}	p / MPa	ρ / kg·m^{-3}	p / MPa	ρ / kg·m^{-3}	p / MPa	ρ / kg·m^{-3}
1.007	1025.3	1.003	1017.6	1.045	1009.9	1.004	1002.0	1.013	994.4	1.005	986.7
1.989	1025.8	2.020	1018.2	2.032	1010.5	1.998	1002.6	2.013	995.0	2.045	987.3
3.027	1026.4	3.006	1018.7	2.995	1011.0	3.050	1003.2	3.004	995.6	2.982	987.8
4.003	1026.9	4.031	1019.2	3.991	1011.6	4.015	1003.7	3.999	996.1	4.026	988.4
5.004	1027.3	5.002	1019.7	4.983	1012.1	4.998	1004.3	5.003	996.8	5.007	988.9
6.004	1027.8	6.020	1020.2	6.016	1012.6	6.020	1004.9	6.019	997.3	6.014	989.5
6.996	1028.3	7.003	1020.7	6.998	1013.2	7.011	1005.4	7.005	997.8	7.015	990.2
8.006	1028.8	8.000	1021.2	8.009	1013.7	8.034	1006.0	8.001	998.4	7.998	990.8
9.066	1029.4	9.006	1021.7	8.993	1014.2	9.001	1006.5	8.984	999.0	9.003	991.3
10.021	1029.8	9.991	1022.2	9.991	1014.7	10.000	1007.1	9.981	999.5	9.986	991.9
11.008	1030.3	11.005	1022.7	10.985	1015.3	11.000	1007.6	11.029	1000.2	10.996	992.5
11.995	1030.8	12.000	1023.2	12.014	1015.8	12.012	1008.2	11.980	1000.7	12.008	993.1
13.024	1031.3	13.010	1023.7	13.079	1016.3	12.947	1008.7	13.051	1001.3	13.004	993.7
13.984	1031.7	14.000	1024.2	13.992	1016.8	13.991	1009.2	14.000	1001.8	13.985	994.2
14.994	1032.2	14.987	1024.7	14.999	1017.3	15.001	1009.8	14.988	1002.4	15.010	994.8
15.992	1032.7	15.987	1025.2	15.997	1017.8	15.987	1010.3	16.018	1002.9	15.983	995.4
16.977	1033.1	17.012	1025.6	16.989	1018.3	16.998	1010.8	16.978	1003.4	17.007	995.7
17.994	1033.6	17.991	1026.1	17.997	1018.8	18.005	1011.3	17.977	1004.0	17.985	996.3
19.000	1034.0	18.995	1026.6	19.011	1019.3	18.981	1011.9	19.003	1004.5	18.994	996.9
19.985	1034.5	20.012	1027.1	19.963	1019.7	19.980	1012.4	19.990	1005.1	19.994	997.4

Table 2. Experimental Liquid Densities, ρ, and Excess Molar Volume, V_m^E, for the MDEA (1) + Water (2) Mixture at $x_1 = 0.0369$

p / MPa	ρ / kg·m^{-3}	V_m^E / cm^3·mol^{-1}	p / MPa	ρ / kg·m^{-3}	V_m^E / cm^3·mol^{-1}	p / MPa	ρ / kg·m^{-3}	V_m^E / cm^3·mol^{-1}	p / MPa	ρ / kg·m^{-3}	V_m^E / cm^3·mol^{-1}	p / MPa	ρ / kg·m^{-3}	V_m^E / cm^3·mol^{-1}
T/K = 313.12			T/K = 323.09			T/K = 333.04			T/K = 352.85			T/K = 362.71		
1.001	1005.7	−0.1438	1.034	1001.1	−0.1502	1.012	995.9	−0.1562	1.004	983.9	−0.1622	1.012	977.3	−0.1654
2.015	1006.1	−0.1423	2.004	1001.5	−0.1493	1.999	996.3	−0.1547	2.037	984.4	−0.1623	2.054	977.8	−0.1651
2.986	1006.5	−0.1415	2.993	1001.9	−0.1483	2.995	996.7	−0.1536	2.962	984.8	−0.1615	3.004	978.2	−0.1635
3.994	1006.9	−0.1402	3.997	1002.4	−0.1492	4.008	997.2	−0.1545	4.014	985.2	−0.1592	3.981	978.6	−0.1622
5.003	1007.3	−0.1390	4.998	1002.7	−0.1459	4.992	997.6	−0.1530	4.994	985.6	−0.1575	4.997	979.1	−0.1623
6.002	1007.7	−0.1379	6.003	1003.1	−0.1444	5.983	998.0	−0.1519	6.034	986.1	−0.1581	6.006	979.5	−0.1603
6.994	1008.1	−0.1369	6.993	1003.5	−0.1434	6.994	998.4	−0.1508	6.999	986.5	−0.1567	7.005	979.9	−0.1584
7.995	1008.5	−0.1358	7.994	1004.0	−0.1445	7.997	998.8	−0.1495	8.019	987.0	−0.1570	7.962	980.3	−0.1568
8.996	1008.9	−0.1348	8.996	1004.4	−0.1434	8.994	999.2	−0.1482	8.987	987.3	−0.1536	8.999	980.8	−0.1568
9.983	1009.3	−0.1342	9.997	1004.8	−0.1425	9.994	999.6	−0.1471	9.992	987.8	−0.1543	9.999	981.2	−0.1554
10.989	1009.7	−0.1331	10.986	1005.2	−0.1414	10.996	1000.0	−0.1460	10.987	988.2	−0.1530	10.997	981.6	−0.1535
11.991	1010.1	−0.1321	11.988	1005.6	−0.1410	11.991	1000.4	−0.1451	11.994	988.6	−0.1513	11.979	982.0	−0.1519
12.991	1010.5	−0.1310	12.997	1006.0	−0.1399	12.996	1000.8	−0.1441	12.978	989.1	−0.1524	12.988	982.5	−0.1527
13.988	1010.9	−0.1306	13.999	1006.4	−0.1389	13.988	1001.2	−0.1432	13.987	989.4	−0.1491	13.990	982.9	−0.1509
14.989	1011.4	−0.1317	14.989	1006.8	−0.1382	14.991	1001.6	−0.1421	14.998	989.9	−0.1496	14.982	983.4	−0.1514
15.985	1011.7	−0.1291	16.002	1007.1	−0.1350	15.994	1002.0	−0.1412	15.956	990.2	−0.1466	15.982	983.8	−0.1501
16.984	1012.1	−0.1280	16.988	1007.6	−0.1362	16.992	1002.4	−0.1403	16.987	990.7	−0.1469	16.992	984.2	−0.1484
17.985	1012.5	−0.1272	17.990	1007.9	−0.1336	17.985	1002.8	−0.1394	17.991	991.1	−0.1458	17.958	984.6	−0.1475
18.989	1012.9	−0.1267	18.996	1008.3	−0.1327	18.990	1003.2	−0.1385	18.986	991.5	−0.1449	18.984	985.1	−0.1478
19.945	1013.3	−0.1260	19.995	1008.7	−0.1319	19.977	1003.6	−0.1378	19.988	991.9	−0.1438	19.984	985.5	−0.1467

simultaneously using a cathetometer with a video camera. This apparatus has already been described and tested in previous works.[28−30] A sapphire tube cell is connected to a vibrating tube densimeter (VTD, Anton Parr DMA 60/512P) for feeding and pressure control purposes. The temperature of the vibrating tube densimeter is thermoregulated by a constant liquid bath using water as thermal fluid, and the temperature of the feeding cell is regulated using an air bath. The temperature stabilities of the liquid bath and air bath are maintained within (± 0.01 and ± 0.05) K, respectively.

The temperature was measured with three platinum probes (Specitec, France), connected to a digital indicator (Automatic Systems F250, USA), which were calibrated against a calibration system (Automatic System F300S, USA), connected to a 25-Ω reference probe (model 162 CE from Rosemount, England, with ± 0.005 K certified accuracy traceable to the ITS-90 protocol). The experimental uncertainty in temperature measurements is estimated to be ± 0.03 K. The pressure was measured with a 25 MPa pressure transducer (BOURDON SEDEME type TF01 250, France) connected to a 6.5 digital multimeter (HP-34401A, USA), which was calibrated at temperatures from (313 to 363) K against a dead weight balance (Desgranges & Hout, model 5304, accuracy ± 0.005 % full scale, France). The uncertainty in pressure measurements was estimated to be ± 0.008 MPa, and the reproducibility of pressure measurements was better than 0.01 %. Both calibration procedures are given elsewhere by Galicia-Luna et al.[31]

Density determinations are based on the measuring of the period of oscillation of the vibrating U-shaped tube, which is filled with the sample liquid. The vibrating tube densimeter is calibrated with two reference fluids. Calibration of the VTD covers the whole range of pressures and temperatures of the measurements performed in this work. The calibrations of the VTD, temperature probes, and pressure transducer are made before and after a new system is studied. In this work, nitrogen and water were used following the classical method.[27] Reference density values of nitrogen and water are obtained with the equations of state of Span et al.[32] and Wagner and Pruss,[33] respectively.

Table 3. Experimental Liquid Densities, ρ, and Excess Molar Volume, V_m^E, for the MDEA (1) + Water (2) Mixture at $x_1 = 0.0607$

p/MPa	ρ/kg·m^{-3}	V_m^E/cm^3·mol^{-1}	p/MPa	ρ/kg·m^{-3}	V_m^E/cm^3·mol^{-1}	p/MPa	ρ/kg·m^{-3}	V_m^E/cm^3·mol^{-1}	p/MPa	ρ/kg·m^{-3}	V_m^E/cm^3·mol^{-1}
	T/K = 313.14			*T*/K = 323.08			*T*/K = 333.03			*T*/K = 342.96	
0.999	1018.1	−0.377	1.011	1012.5	−0.371	1.015	1006.4	−0.365	1.042	999.5	−0.348
2.000	1018.5	−0.375	2.015	1012.9	−0.369	1.998	1006.8	−0.362	2.006	999.8	−0.345
3.028	1018.9	−0.373	3.015	1013.2	−0.365	3.013	1007.2	−0.361	3.028	1000.3	−0.344
4.020	1019.3	−0.372	4.025	1013.7	−0.365	4.037	1007.6	−0.358	4.013	1000.7	45.952
5.011	1019.7	−0.370	4.995	1014.0	−0.361	5.116	1008.1	−0.358	5.003	1001.2	−0.343
6.015	1020.1	−0.369	6.015	1014.4	−0.360	6.004	1008.4	−0.355	5.994	1001.6	−0.341
7.003	1020.4	−0.365	7.015	1014.8	−0.358	6.992	1008.8	−0.354	7.004	1002.0	−0.339
8.009	1020.8	−0.363	7.992	1015.2	−0.357	8.003	1009.1	−0.350	8.063	1002.5	−0.339
9.014	1021.2	−0.363	9.006	1015.6	−0.355	8.975	1009.6	−0.351	9.140	1003.0	−0.339
10.000	1021.5	−0.359	10.014	1016.0	−0.354	9.983	1009.9	−0.346	10.022	1003.4	−0.338
10.977	1022.0	−0.360	11.000	1016.3	−0.350	11.029	1010.4	−0.347	11.032	1003.7	−0.334
12.029	1022.3	−0.356	12.064	1016.8	−0.351	12.015	1010.7	−0.343	11.997	1004.1	−0.333
13.022	1022.7	−0.355	13.004	1017.1	−0.348	13.028	1011.1	−0.342	13.009	1004.5	−0.331
13.988	1023.0	−0.352	13.990	1017.5	−0.347	14.020	1011.5	−0.340	14.001	1004.9	−0.329
15.009	1023.4	−0.350	14.997	1017.9	−0.345	15.022	1011.9	−0.339	15.005	1005.3	−0.328
16.003	1023.8	−0.349	15.999	1018.2	−0.342	16.006	1012.3	−0.338	16.002	1005.7	−0.326
16.995	1024.2	−0.348	16.993	1018.6	−0.340	16.996	1012.7	−0.336	17.011	1006.1	−0.325
18.007	1024.6	−0.347	18.008	1019.0	−0.339	18.013	1013.1	−0.335	17.988	1006.5	−0.323
19.000	1024.9	−0.344	19.017	1019.4	−0.338	19.018	1013.4	−0.331	18.992	1006.9	−0.322
19.950	1025.3	−0.343	19.961	1019.7	−0.335	19.965	1013.8	−0.331	20.154	1007.3	−0.318

Table 4. Experimental Liquid Densities, ρ, and Excess Molar Volume, V_m^E, for the MDEA (1) + Water (2) Mixture at $x_1 = 0.0893$

p/MPa	ρ/kg·m^{-3}	V_m^E/cm^3·mol^{-1}	p/MPa	ρ/kg·m^{-3}	V_m^E/cm^3·mol^{-1}	p/MPa	ρ/kg·m^{-3}	V_m^E/cm^3·mol^{-1}
	T/K = 313.13			*T*/K = 323.10			*T*/K = 333.05	
1.011	1025.9	−0.542	1.009	1019.6	−0.528	1.013	1013.0	−0.517
1.994	1026.2	−0.538	2.006	1020.0	−0.526	2.006	1013.4	−0.514
3.002	1026.6	−0.536	3.012	1020.4	−0.524	2.994	1013.8	−0.512
3.998	1027.0	−0.534	4.011	1020.8	−0.521	4.031	1014.2	−0.509
4.998	1027.4	−0.532	5.002	1021.2	−0.520	4.999	1014.6	−0.506
6.001	1027.8	−0.530	5.996	1021.6	−0.518	6.000	1015.0	−0.505
6.994	1028.1	−0.525	7.006	1022.0	−0.515	6.995	1015.4	−0.503
8.000	1028.5	−0.523	8.004	1022.3	−0.510	8.005	1015.8	−0.501
8.999	1028.9	−0.522	8.991	1022.7	−0.508	9.000	1016.2	−0.499
9.995	1029.2	−0.518	9.999	1023.1	−0.506	9.992	1016.5	−0.493
10.990	1029.6	−0.516	10.993	1023.5	−0.506	10.993	1017.0	−0.494
12.001	1030.0	−0.514	11.992	1023.8	−0.501	11.998	1017.3	−0.489
12.989	1030.3	−0.511	13.001	1024.2	−0.499	13.000	1017.7	−0.487
14.000	1030.7	−0.509	13.990	1024.6	−0.498	14.008	1018.1	−0.486
14.999	1031.1	−0.507	14.980	1025.0	−0.496	14.983	1018.5	−0.484
15.995	1031.4	−0.504	16.002	1025.3	−0.491	16.002	1018.9	−0.482
17.013	1031.8	−0.502	16.990	1025.7	−0.490	17.032	1019.3	−0.480
17.993	1032.2	−0.500	17.994	1026.1	−0.489	17.945	1019.6	−0.476
18.991	1032.6	−0.499	18.999	1026.5	−0.487	18.989	1020.0	−0.474
19.991	1032.9	−0.495	20.002	1026.8	−0.483	19.994	1020.4	−0.473
	T/K = 342.94			*T*/K = 352.88			*T*/K = 362.75	
1.029	1005.9	−0.500	1.037	998.6	−0.489	0.993	990.9	−0.476
1.996	1006.3	−0.498	2.010	999.0	−0.487	2.025	991.3	−0.471
3.018	1006.7	−0.495	3.005	999.5	−0.486	3.013	991.8	−0.472
3.995	1007.1	−0.493	4.010	999.9	−0.483	3.995	992.2	−0.468
5.000	1007.5	−0.490	4.999	1000.3	−0.481	5.018	992.7	−0.467
5.992	1007.9	−0.488	5.993	1000.7	−0.478	6.003	993.1	−0.464
7.008	1008.3	−0.485	7.000	1001.1	−0.475	6.988	993.6	−0.463
7.994	1008.8	−0.486	7.998	1001.6	−0.475	8.002	994.0	−0.460
9.001	1009.1	−0.480	8.982	1002.0	−0.472	8.996	994.4	−0.458
9.994	1009.6	−0.481	9.977	1002.4	−0.470	10.040	994.9	−0.457
10.983	1010.0	−0.479	10.995	1002.8	−0.467	11.000	995.3	−0.454
12.000	1010.4	−0.477	11.987	1003.3	−0.467	11.988	995.7	−0.452
12.996	1010.8	−0.474	12.999	1003.7	−0.464	12.992	996.1	−0.448
14.014	1011.2	−0.472	13.990	1004.1	−0.462	13.990	996.6	−0.448
14.994	1011.6	−0.470	14.969	1004.5	−0.459	14.990	997.0	−0.446
15.989	1012.0	−0.468	15.990	1004.9	−0.457	15.993	997.5	−0.445
16.991	1012.4	−0.465	16.991	1005.3	−0.455	17.004	997.9	−0.443
17.989	1012.8	−0.464	18.009	1005.8	−0.455	17.984	998.3	−0.440
18.984	1013.2	−0.462	18.994	1006.2	−0.453	19.022	998.7	−0.437
19.988	1013.6	−0.460	20.015	1006.6	−0.451	19.984	999.2	−0.438

Procedure. This consists of four steps. First, temperature and pressure sensors and the densimeter are calibrated. Second, the liquid sample is prepared and introduced into the feeding cell. Third, experimental conditions are set up in the equipment.

Table 5. Experimental Liquid Densities, ρ, and Excess Molar Volume, V_m^E, for the MDEA (1) + Water (2) Mixture at $x_1 = 0.1302$

p / MPa	ρ / kg·m^{-3}	V_m^E / cm^3·mol^{-1}	p / MPa	ρ / kg·m^{-3}	V_m^E / cm^3·mol^{-1}	p / MPa	ρ / kg·m^{-3}	V_m^E / cm^3·mol^{-1}
	T/K = 313.13			T/K = 323.09			T/K = 333.03	
1.003	1033.5	−0.745	1.017	1026.6	−0.723	1.009	1019.4	−0.703
1.993	1033.9	−0.742	2.001	1027.0	−0.720	2.002	1019.8	−0.699
2.999	1034.3	−0.739	3.015	1027.4	−0.717	2.997	1020.2	−0.697
3.994	1034.6	−0.734	4.003	1027.8	−0.715	3.993	1020.6	−0.694
4.984	1035.0	−0.731	4.999	1028.1	−0.709	4.997	1021.0	−0.690
6.001	1035.4	−0.728	5.998	1028.5	−0.705	6.002	1021.4	−0.687
6.993	1035.8	−0.726	6.998	1028.9	−0.703	6.992	1021.8	−0.685
8.003	1036.1	−0.721	7.999	1029.3	−0.700	8.013	1022.2	−0.680
8.996	1036.5	−0.720	8.998	1029.7	−0.698	9.016	1022.6	−0.678
9.989	1036.8	−0.714	9.995	1030.1	−0.695	9.993	1023.0	−0.676
10.983	1037.2	−0.712	10.997	1030.4	−0.690	10.989	1023.4	−0.673
11.992	1037.6	−0.709	11.998	1030.8	−0.689	12.007	1023.8	−0.671
12.980	1037.9	−0.705	13.005	1031.2	−0.687	13.003	1024.1	−0.665
14.000	1038.3	−0.703	14.003	1031.6	−0.684	13.995	1024.5	−0.663
15.002	1038.7	−0.700	14.990	1032.0	−0.682	15.011	1024.9	−0.660
16.001	1039.1	−0.699	15.996	1032.3	−0.677	15.996	1025.3	−0.658
16.998	1039.5	−0.697	16.999	1032.7	−0.674	17.011	1025.7	−0.656
17.991	1039.8	−0.692	17.996	1033.1	−0.672	17.985	1026.1	−0.654
18.996	1040.2	−0.691	18.988	1033.4	−0.668	18.991	1026.4	−0.648
19.990	1040.5	−0.686	20.004	1033.8	−0.666	19.993	1026.8	−0.646
	T/K = 342.91			T/K = 352.84			T/K = 362.72	
1.008	1011.7	−0.676	1.009	1004.0	−0.658	1.043	995.9	−0.635
1.996	1012.1	−0.673	2.026	1004.3	−0.651	2.117	996.4	−0.633
3.044	1012.5	−0.668	3.005	1004.8	−0.651	3.008	996.8	−0.629
4.015	1012.9	−0.664	4.006	1005.3	−0.650	3.995	997.3	−0.629
5.006	1013.4	−0.665	5.004	1005.7	−0.645	5.002	997.8	−0.628
5.995	1013.8	−0.662	5.993	1006.1	−0.643	5.994	998.2	−0.624
7.010	1014.2	−0.658	6.991	1006.5	−0.638	6.994	998.6	−0.619
8.001	1014.6	−0.655	7.992	1006.9	−0.634	8.003	999.1	−0.618
8.996	1015.0	−0.651	8.994	1007.4	−0.634	8.988	999.6	−0.616
9.995	1015.5	−0.652	9.980	1007.8	−0.630	9.991	1000.0	−0.614
10.992	1015.9	−0.649	10.971	1008.2	−0.628	11.003	1000.4	−0.609
12.029	1016.3	−0.645	11.987	1008.7	−0.626	12.036	1000.9	−0.608
12.993	1016.7	−0.643	13.040	1009.1	−0.623	13.005	1001.3	−0.605
13.996	1017.1	−0.640	13.997	1009.5	−0.620	13.983	1001.7	−0.601
14.997	1017.5	−0.638	14.981	1009.9	−0.617	14.984	1002.2	−0.601
15.998	1017.9	−0.634	15.989	1010.4	−0.618	15.981	1002.6	−0.597
16.997	1018.3	−0.631	16.976	1010.8	−0.613	16.993	1003.0	−0.593
18.033	1018.8	−0.632	17.977	1011.2	−0.611	17.988	1003.5	−0.593
18.977	1019.2	−0.630	18.979	1011.6	−0.608	18.994	1003.9	−0.589
19.980	1019.6	−0.627	19.995	1012.0	−0.604	19.995	1004.4	−0.589

Finally, measurements are made at equilibrium. Four measurements of the same point are recorded, once the pressure and period signals are kept constant.

The density of the fluid inside the vibrating tube, ρ_F, is calculated by

$$\rho_F(p,T) = \rho_{H_2O}(p,T) + \frac{1}{A}(p,T)(\tau^2{}_F(p,T) - \tau^2{}_{H_2O}(p,T)) \quad (1)$$

$$\frac{1}{A}(p,T) = \frac{\rho_{H_2O}(p,T) - \rho_{N_2}(p,T)}{\tau^2{}_{H_2O}(p,T) - \tau^2{}_{N_2}(p,T)} \quad (2)$$

where $\rho_F(p,T)$, $\rho_{H_2O}(p,T)$, and $\rho_{N_2}(p,T)$ are the densities of the liquid sample to be studied, water, and nitrogen, respectively. Besides $\tau_F(p,T)$, $\tau_{H_2O}(p,T)$ and $\tau_{N_2}(p,T)$ are the periods of oscillation of the fluid, water, and nitrogen, respectively. The uncertainty of the density measurements was obtained from the law of propagation of errors[34] using eqs 1 and 2 as previously reported.[35] The uncertainty in density measurements is estimated to be ± 0.2 kg·m^{-3} at a 95 % confidence level.

Loading of the Measurement Cell. The complete procedure is presented in a precedent paper.[27] Samples with the desired compositions are prepared by successive loadings of the pure compounds in the sapphire feeding cell with a maximum volume of 12 cm^3. The amounts of the compounds are determined by weightings carried out within ± 10^{-7} kg accuracy using a comparator balance (Sartorius MCA1200, Germany), which is periodically calibrated with a standard mass of 1 kg class E1. The resulting uncertainty for the mole fraction compositions of the mixtures is lower than ± 10^{-4}, assuming perfect mixing of the mixture. During experimental measurements, the mixture under study was mixed with the help of a magnetic rod and a variable-speed velocity engine[28−30] to accomplish perfect mixing of the mixture.

Results and Discussion

Density Measurements. The results of the experimental density measurements of MDEA are reported in Table 1. Densities of four gravimetrically prepared mixtures at the following molar fraction compositions, $x_1 = 0.0369$, 0.0607, 0.0893, and 0.1302, were measured, and the results are reported in Tables 2 to 5. Densities of MDEA and of aqueous solutions of MDEA are plotted in Figure 1 as a function of pressure at 313.12 K.

The densities of MDEA (1) + water (2) at $x_1 = 0.0369$ and 0.0607 are lower than those of MDEA densities, and densities of the mixture at $x_1 = 0.0893$ are similar to those of MDEA. On the other hand, densities of the mixture at $x_1 = 0.1302$ are larger than MDEA densities. This crossover behavior is also presented at atmospheric pressure.[9]

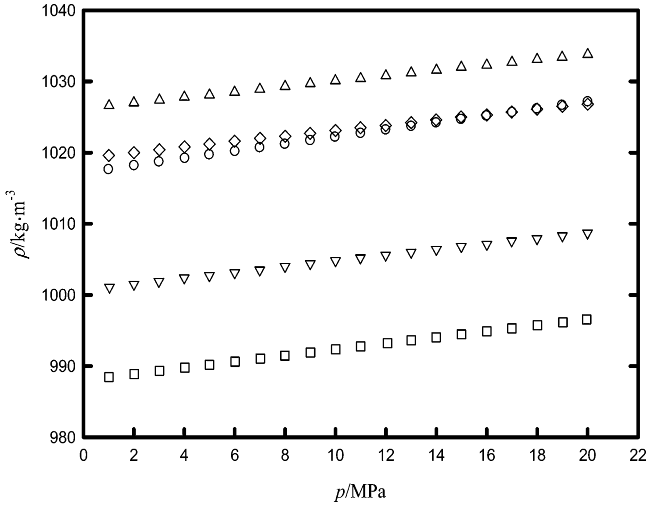

Figure 1. Experimental densities of MDEA and of the MDEA (1) + water (2) binary system at 313.12 K: ○, MDEA; ▽, $x_1 = 0.0369$; □, $x_1 = 0.0607$; ◇, $x_1 = 0.0893$; △, $x_1 = 0.1302$.

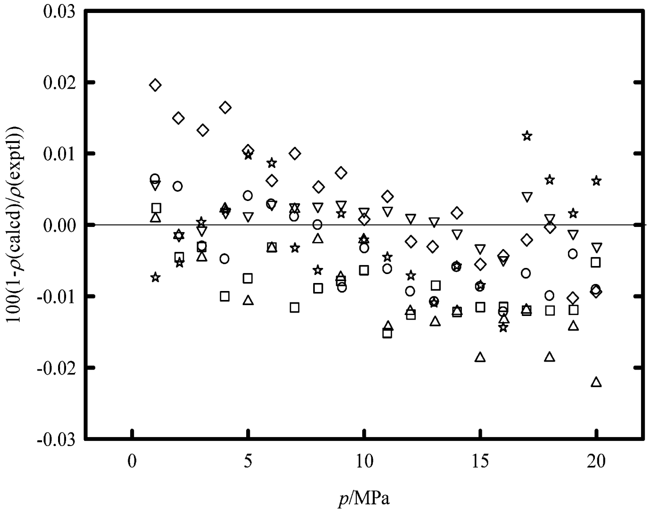

Figure 2. Relative deviations of experimental densities from this work (ρ(exptl)) and values calculated (ρ(calcd)) with the six-parameter equation using the parameters reported in Table 6 for MDEA, at the following temperatures: ○, 313.09 K; ▽, 323.01 K; □, 332.95 K; ◇, 342.78 K; △, 352.68 K; ☆, 362.52 K.

Densities of MDEA and of each mixture were correlated using a volume explicit equation of six parameters,[36] which is a modification of the equations reported by Toscani and Szwarc.[37]

$$v/\text{m}^3\cdot\text{kg}^{-1} = \frac{d_1 + d_2 p}{d_3 - d_4 T + d_5 T^{1/2} + d_6 p} \tag{3}$$

where d_i parameters are reported in Table 6 and were obtained using a Marquardt−Levenberg least-squares optimization procedure using the following objective function, S

$$S = \sum_{i=1}^{n} \left[\frac{\rho_i(\text{exptl}) - \rho_i(\text{calcd})}{\rho_i(\text{exptl})} \right]^2 \tag{4}$$

Temperature, pressure, and density ranges, the number of data points used to obtain the optimized parameters for MDEA and for each mixture, along with statistical values are reported in Table 6. The equations of statistical values to evaluate the correlations have been given previously.[35]

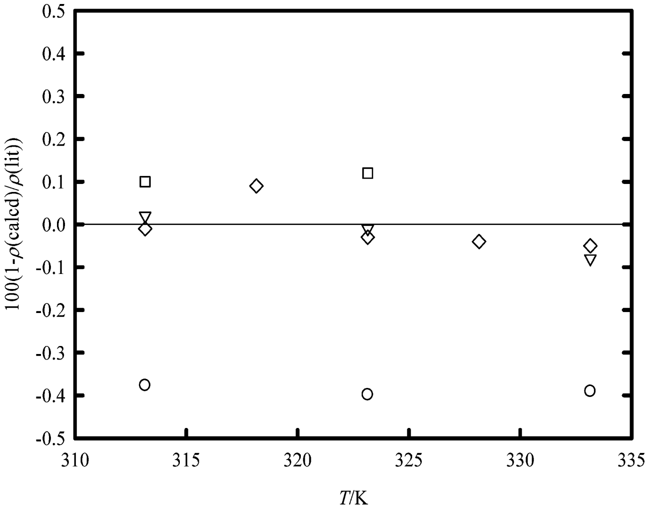

Figure 3. Relative deviations of experimental densities at atmospheric pressure reported by Al-Ghawas et al.[6] (ρ(lit)) and values calculated (ρ(calcd)) with the six-parameter equation using the parameters reported in Table 6 for MDEA (1) + water (2), at the following mole fraction compositions: ○, 0.0369; ▽, 0.0607; □, 0.0893 K; ◇, 0.1302.

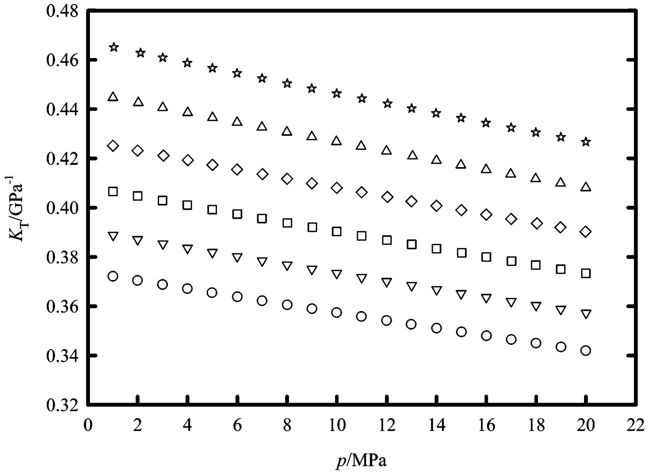

Figure 4. Isotherms for the isothermal compressibilities of the MDEA (1) + water (2) binary mixture at $x_1 = 0.1302$, at the following temperatures: ○, 313.13 K; ▽, 323.09 K; □, 333.03 K; ◇, 342.91 K; △, 352.84 K; ☆, 362.72 K.

Relative deviations of experimental densities of MDEA (ρ(exptl)) and calculated values using the six-parameter equation (ρ(calcd)) are plotted in Figure 2. The maximum deviations are +0.020 % and −0.022 %. Similar plots can be found for the correlations of each mixture. The six-parameter equation represents the experimental densities of MDEA and of each mixture with a standard deviation better than 0.01 %.

Some experimental data reported by Hawrylak et al.[12] were compared with values calculated with the correlations obtained in this work. This comparison was made to check the consistency of our correlations at high pressure. The comparisons are reported in Table 7 for mixtures of similar MDEA composition. The data points compared agree within an average absolute deviation (AAD) of 0.25 %. However, there are slight differences in the composition of MDEA in the aqueous solutions between data of Hawrylak et al.[12] and data from this work. The data with closer compositions ($x_1 = 0.0605$[12] and 0.0607) were found to have less deviation, whereas for a difference in composition of 0.0006 ($x_1 = 0.0375$[12] and 0.0369), there was an AAD of 0.26 %. Therefore, the difference in composition has some influence on the deviation of the densities compared.

Table 6. Ranges of Temperature, T, Pressure, p, and Density, ρ, Data Points, n, and Parameters for the Correlation Model for MDEA and for Mixtures of MDEA (1) + Water (2) along with Statistical Values: Average Absolute Deviation (AAD), Mean Deviation, Bias, Standard Deviation (SDV), and Root-Mean-Square (rms)

	MDEA	$x_1 = 0.0369$	$x_1 = 0.0607$	$x_1 = 0.0893$	$x_1 = 0.1302$
T_{min}/K	313.09	313.12	313.14	313.13	313.13
T_{max}/K	362.52	362.71	342.96	362.75	362.72
p_{min}/MPa	1.003	1.001	0.999	0.993	1.003
p_{max}/MPa	20.012	19.995	20.154	20.015	20.004
ρ_{min}/kg·m^{-3}	986.7	977.3	999.5	990.9	995.9
ρ_{max}/kg·m^{-3}	1034.5	1013.3	1025.3	1032.9	1040.5
n	120	100	80	120	120
d_1/MPa·m^3·kg^{-1}	−1.839	−2.002	−2.570	−5.311	−2.436
d_2/m^3·kg^{-1}	−0.00469	−0.00225	−0.00502	−0.01048	−0.00516
d_3/MPa	−2193.1	38.7	37.4	−2200.8	−1513.7
d_4/MPa·K^{-1}	−1.872	−8.303	−11.236	−16.872	−6.551
d_5/MPa·K$^{-1/2}$	−15.69	−262.85	−348.75	−481.97	−172.60
d_6	−5.74	−3.08	−6.11	−12.79	−6.27
AAD/%	0.006	0.004	0.004	0.004	0.005
bias/%	−0.003	0.002	−0.001	−0.001	−0.003
SDV/%	0.008	0.004	0.006	0.005	0.006
rms/%	0.008	0.005	0.006	0.005	0.007

Table 7. Comparisons of Density Data of the MDEA (1) + Water (2) System Reported by Hawrylak et al.[12] with Values Calculated with the Correlations Obtained in This Work

T	p	x_1		ρ/kg·m^{-3}		$100(\rho(\text{lit}) - \rho(\text{calcd}))$
K	MPa	Hawrylaket al.[12]	this work	Hawrylaket al.[12]	this work	ρ(lit)
283.15	0.1	0.0375	0.0369	1020.0	1015.7	0.42
298.15	0.1	0.0375	0.0369	1015.1	1011.2	0.38
313.15	0.1	0.0375	0.0369	1008.6	1005.3	0.32
328.15	0.1	0.0375	0.0369	1000.8	998.1	0.27
337.46	10.195	0.0375	0.0369	999.9	997.2	0.27
337.50	10.189	0.0605	0.0607	1006.6	1007.1	−0.05
335.16	20.198	0.0375	0.0369	1004.2	1002.6	0.16
335.24	20.194	0.0605	0.0607	1011.4	1012.5	−0.10

The correlations obtained in this work were extrapolated to atmospheric pressure and compared with some experimental densities reported in the literature[2,6,8] to check for the capability of the correlations obtained here. The aqueous mixtures reported in the literature are at $w_1 = 0.2$, 0.3, 0.4, and 0.5, and these correspond on a fraction mole basis to 0.0364, 0.0608, 0.0916, and 0.1313, respectively. The comparisons were made in the range of temperatures measured here. For MDEA, the calculated values and the literature values[2,8] agree with an AAD of 0.05 %. For mixtures only, the data reported by Al-Ghawas et al.[6] were used to perform comparisons, and the relative deviations are reported in Figure 3. For a mixture at $x_1 = 0.0369$, the AAD between literature data[6] and calculated ones was 0.3 %. For a mixture at $x_1 = 0.0607$, the AAD between literature data[6] and calculated values was 0.04 %. For the case of a mixture at $x_1 = 0.0893$, the AAD was 0.08 %. For a mixture at $x_1 = 0.1302$, the density data reported[6] and the calculated values are in agreement within an AAD of 0.04 %.

The density correlations obtained for MDEA and for mixtures at $x_1 = 0.0607$, 0.0893, and 0.1302 are in good agreement with data reported in the literature, and the existing differences can be attributable to the small differences in the composition of the mixture and combined uncertainties of both sets of data. For the correlation at $x_1 = 0.0369$, the AAD is higher (0.3 %) than those obtained for the remaining mixtures studied in this work and the differences in composition are not large; however, the densities reported by Al-Ghawas et al.[6] at $x_1 = 0.0364$ follow the same behavior as those reported by Hawrylak et al.[12] at $x_1 = 0.0375$. Evermore, the magnitude of densities reported by Al-Ghawas et al.[6] is larger than those reported by Hawrylak et al.,[12] and therefore the deviations with densities reported by Al-Ghawas et al.[6] at $x_1 = 0.0364$ are higher.

Excess Molar Volumes. The excess molar volumes were calculated according to the relation

$$V_m^E/\text{cm}^3\cdot\text{mol}^{-1} = \frac{x_1 W_1 + x_2 W_2}{\rho^{mix}} - (x_1 V_1 + x_2 V_2) \qquad (5)$$

where V_m^E is the excess molar volume; ρ^{mix} is the density of the mixture; V_1 and V_2 are the pure component molar volumes at the measured temperature and pressure of the mixture; W_1 and W_2 are the molecular weights of MDEA and water, respectively; and x_1 and x_2 are the mole fractions of MDEA and water, respectively. V_1 is calculated using the six-parameter equation, and V_2 is calculated using the EoS proposed by Wagner and Pruss.[33] The uncertainty was estimated to be ± 0.006 cm^3·mol^{-1} for the excess molar volumes in the whole interval of measurements reported in this work. The excess molar volumes are reported in Tables 2 to 5. V_m^E exhibits negative deviations from ideality at the investigated temperature and pressure ranges studied here. V_m^E values are more negative when the temperature increases at constant pressure, and V_m^E values are less negative when the pressure increases at constant temperature.

Derived Thermodynamic Properties. The isothermal compressibility (K_T) and the isobaric thermal expansivity (a_p) can be obtained from density data using the following expressions

$$K_T = \frac{1}{\rho}\left(\frac{\partial\rho}{\partial p}\right)_T \qquad (6)$$

$$\alpha_p = -\frac{1}{\rho}\left(\frac{\partial\rho}{\partial T}\right)_p \qquad (7)$$

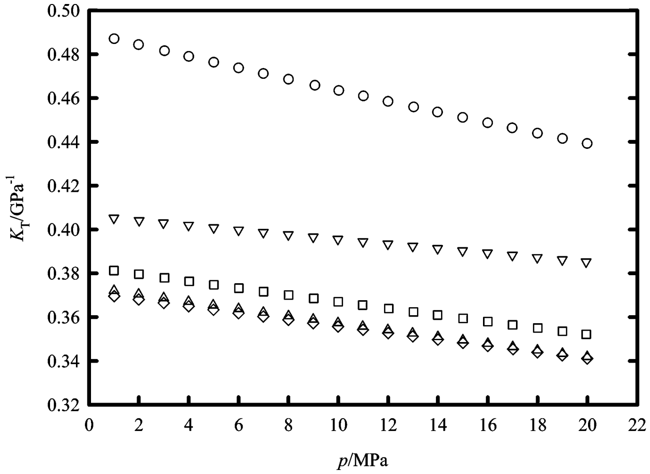

Figure 5. Isothermal compressibilities for MDEA and for the MDEA (1) + water (2) system at 313.12 K: ○, MDEA; ▽, $x_1 = 0.0369$; □, $x_1 = 0.0607$; ◇, $x_1 = 0.0893$; △, $x_1 = 0.1302$.

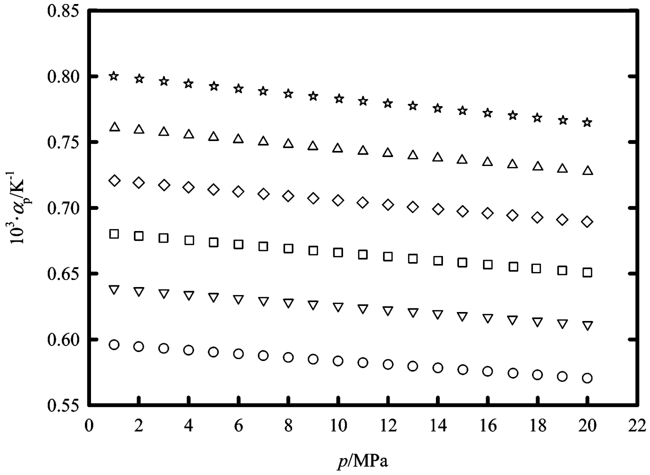

Figure 6. Isotherms for the isobaric thermal expansivity for the MDEA (1) + water (2) binary mixture at $x_1 = 0.0893$, at the following temperatures: ○, 313.13 K; ▽, 323.10 K; □, 333.05 K; ◇, 342.94 K; △, 352.88 K; ☆, 362.75 K.

In this work, the six-parameter equation was substituted in eqs 6 and 7; therefore, K_T and a_p are calculated using the following two expressions[36]

$$K_T/\text{GPa}^{-1} = 1000[d_6/(d_3 - d_4T + d_5T^{1/2} + d_6p) - d_2/(d_1 + d_2p)] \tag{8}$$

$$\alpha_p/\text{K}^{-1} = (2d_4 - d_5T^{-1/2})/(2(d_3 - d_4T + d_5T^{1/2} + d_6p)) \tag{9}$$

The uncertainties of these two thermodynamic derived properties were obtained on the basis of the parameters obtained for the six-parameter equation and were obtained according to the procedure presented in a previous work.[36] The uncertainties were estimated to be ± 0.005 GPa^{-1} and ± $5 \cdot 10^{-7}$ K^{-1} for K_T and α_p, respectively.

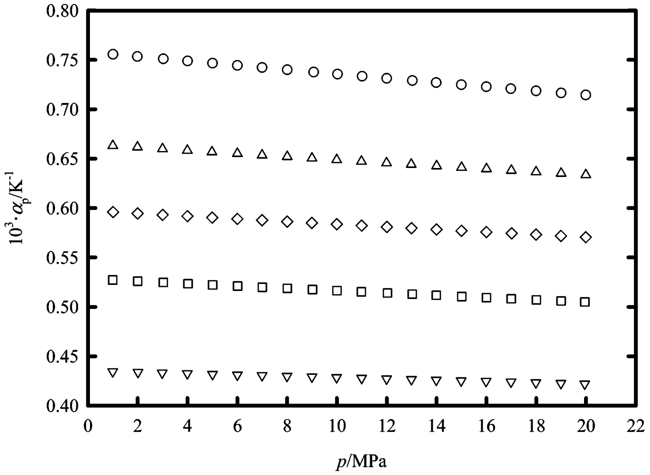

Figure 7. Isobaric thermal expansivities for MDEA and for the MDEA (1) + water (2) system at 313.12 K: ○, MDEA; ▽, $x_1 = 0.0369$; □, $x_1 = 0.0607$; ◇, $x_1 = 0.0893$; △, $x_1 = 0.1302$.

To check for the capability of the six-parameter equation to represent K_T and α_p, the data reported by Fandiño et al.[38] for pentaerythritol ester lubricant mixtures were calculated. First, densities of three different systems reported by Fandiño et al.[37] were correlated with eq 3. These densities have an order of magnitude similar to that reported in this work. Then, K_T and α_p were calculated and compared with the reference values.[37] The average absolute deviations between reference values[37] and calculated values were 1.4 % and 0.6 % for K_T and α_p, respectively.

The calculated K_T and α_p values for MDEA and for each mixture are reported in a PDF file as Supporting Information (Tables SI-1 to SI-5). K_T decreases as the pressure increased; on the other hand, K_T increases as the temperature increased, as can be observed in Figure 4 for a mixture of MDEA (1) + water (2) at $x_1 = 0.1302$. The isothermal compressibilities as function of pressure for MDEA and for each mixture at 313.12 K are plotted in Figure 5. K_T values for MDEA are larger than those for the mixtures studied here. However, K_T decreases as the composition of MDEA in the mixture increases to reach a mole fraction composition of 0.0893, and then the K_T increases for a mixture at $x_1 = 0.1302$.

The isobaric thermal expansivities decrease as the pressure decreases at constant temperature, and α_p increases as the temperature increases at constant pressure. These effects are illustrated in Figure 6 for a mixture of MDEA (1) + water (2) at $x_1 = 0.0893$. The isobaric thermal expansivities as a function of pressure for MDEA and for each mixture at 313.12 K are plotted in Figure 7. α_p increases as the composition of MDEA in the mixture is increased.

Conclusions

The densities of MDEA and of the MDEA + water binary mixture were measured at high pressure in this work. Measurements were performed at temperatures from (313 to 363) K and pressures up to 20 MPa. The data reported here complement the data reported by Hawrylak et al.[12] Four binary mixtures at $x_1 = 0.0369$, 0.0607, 0.0893, and 0.1302 were studied. The experimental density data were correlated with a short explicit volume equation of six parameters. This equation correlated the densities of MDEA and of MDEA + water with a standard deviation better than 0.01 %. Densities from the literature were compared with values calculated with the correlations obtained. Good agreement was found with the density data at atmospheric and high pressure with data reported by Hawrylak et al.[12] At atmospheric pressure, the data reported by Al-Ghawas et al.[6] showed good agreement with our correlations. Only for the mixture at $w_1 = 0.2$, there were some deviations, but comparing data of Al-Ghawas and Hawrylak et al.,[12] we conclude that the data of Al-Ghawas are at a slightly higher MDEA composition. The excess molar volumes were calculated for the binary mixtures, and all showed negative deviations from ideality in the range of measurements performed in this work. Isothermal compressibilities and isobaric thermal expansivities were calculated using the six-parameter equation.

Supporting Information Available:

Calculated isothermal compressibilities and isobaric thermal expansivities for the systems studied here. This material is available free of charge via the Internet at http://pubs.acs.org.

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