532.13:536.22

$Ca(NO_3)_2$ (aq) AS HEAT TRANSFER FLUID FOR THE ALTERNATIVE ENERGY RESOURCES. I. (p, ρ, T) and (p_S, ρ_S, T_S) PROPERTIES

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(p, ρ, T) and (p_s, ρ_s, T_s) properties of $Ca(NO_3)_2$ (aq) at T=(298.15 to 398.15) K, at pressures up to p=60 MPa were reported. The experiments were carried out at molalities m=(0.18848, 0.32075, 0.52994, 1.07546, 2.03143, and 3.28155) mol·kg^{-1} of calcium nitrate.

The absorption refrigerating machines and absorption heat pumps successful are used for the receiving of cold and heat as alternative energy resources. For the analysis of detailed properties of these systems, it is necessary to know the thermodynamic properties of refrigeration agents and heat transfer rates of these installations [1]. The serious problems in using the conventional LiBr + H_2O solution as a working fluid are crystallization and corrosion, because the high absorber temperature caused by the air-cooling needs a higher absorbent concentration of the working fluid to maintain low vapor pressure in the absorber. The heat transfer fluids of these systems will be thermal stability during all working range of temperatures. The reaction with metals, corrosion, consolidation and crystallization materials should be minimal. One of the common methods to reduce crystallization of the working fluid is to add a small amount of other salts and organic substance having nonvolatile and hygroscopic properties have been used. Solutions for these systems widely apply various anticorrosive and anticristallisation additivities, such as calcium nitrate [2]. Previous investigations [3-22] of the (p, ρ, T) and (p_s, ρ_s, T_s) properties of $Ca(NO_3)_2$ (aq) are tabulated in Table 1. From the table it is to be seen, that mainly all papers include the results only in ambient pressure, except [18-19]. However, it is necessary to investigate these solutions over a wider range of state parameters in order to provide accurate data for the absorption cycle.

This paper is a continuation of our previous publications [23]. In the present paper, the (p, ρ, T) and (p_s, ρ_s, T_s) properties of $Ca(NO_3)_2$ (aq) at T=(298.15 to 398.15) K, at pressures up to p=60 MPa are reported.

2. Experimental

The (p, ρ, T) and (p_s, ρ_s, T_s) properties were investigated in an experimental installation implementing the constant-volume piezometer method [24]. The apparatus enables the determination of (p, ρ, T) and (p_s, ρ_s, T_s) with high accuracy, as well as the determination of experimental isotherms, isochores, and isobars.

The main part of the installation is a spherical, thick-walled, high-pressure vessel manufactured of 1Cr18Ni9Ti grade stainless steel. The volume of piezometer (0.35013 dm^3) was determined by the mass of water filling it at an exactly measured temperature and moderate pressure of the order of 1 to 1.5 MPa.

Under these conditions, the density of ordinary water is known with high accuracy (0.001-0.003%) from the IAPWS formulation values for ordinary water [25]. The reliability of the data obtained was verified after each run by a control measurement of volumetric properties of water. Deviations of all the parameters did not exceed the tolerance given by [25].

TABLE 1

A literature revision of experimental works dedicated to the thermodynamic properties of $Ca(NO_3)_2$ (aq)

First author	Reference	Year	Method	Properties	Uncertainty	Temperature, T/K	Pressure, p/MPa	Concentration
Mylius	[3]	1897		ρ		291.15	0.1	w=0.548
Jones	[4]	1904		ρ		248.83 to 272.95	0.1	w=0.069 to 0.3998
ICT[a]	[5]	1928		ρ		279.15 to 303.15	0.1	w=0.02 to 0.68
Pearce	[6]	1935	OP	ρ, P, a_s, γ		298.15	0.1	m=0 to 8.3601 mol·kg^{-1}
Scott	[7]	1936	P	ρ, α, V_ϕ	0.01 %	308.15	0.1	w=0 to 0.7551
Lamberg	[8]	1948		ρ		273.15 to 313.15	0.1	w=0.48 to 0.643
Ewing	[9]	1950	P	ρ, V_ϕ	0.02 %	298.15 to 333.15	0.1	m=0 to 20.19 mol·kg^{-1}
Rodyanskiy	[10]	1962	A	v		298.15 to 613.15	0.1	m=0.25 to 3.0 mol·kg^{-1}
Romankov	[11]	1969	P	ρ		298.15 to 353.15	0.1	w=0.507 to 0.73
Balashova	[12]	1969		ρ		298.15	0.1	w=0.5763
Millero	[13]	1972						
Blinova	[14]	1978	Review	ρ, η		273.15 to 393.15	0.1	w=0 to 0.7
Spitzer	[15]	1979	FD	V_ϕ	0.5 cm^3·mol^{-1}	298.15	0.1	m=0.03825 to 0.23711 mol·kg^{-1}
Kuznetsov	[16]	1983	P	ρ	±2·10^{-4} g/cm^3	288.15 to 318.15	0.1	m=0.693 to 5.555 mol·kg^{-1}
Jain	[17]	1984	MD	ρ, α, V_ϕ	±0.05 %	293.15 to 333.15	0.1	m=0.0093 to 18.6 mol·kg^{-1}
Akhundov	[18]	1989	CVP	p, ρ, T, w	±0.065 %	298.15 to 598.15	0.1 to 50	w=0.3 to 0.41
Akhundov	[19]	1989	CVP	p, ρ, T, w	±0.065 %	298.15 to 598.15	0.1 to 50	w=0.042 to 0.2
Krumgalz	[20]	1996	Review	V_ϕ	±0.29·10^{-4} g/cm^3	298.15	0.1	
Jahagidar	[21]	1997	P	ρ	±0.0002 g/cm^3	298.15 to 318.15	0.1	m=0.01 to 1.0 mol·kg^{-1}
Vercher	[22]	1999	OTD	ρ, V_ϕ	±0.08 kg/m^3	298.15	0.1	x=0.00507 to 0.1069

[a] ICT, International critical tables; OP, Ostwald pycnometer; P, Pycnometer; A, autocluve; FD, flow densimeter; MD, manometric densimeter; CVP, constant volume piezometer; OTD, oscillating-tube densimeter; ρ, density; P, vapor pressure; a_s, activity of solvent; γ, activity coefficient; α, thermal expansivity; V_ϕ, apparent molar volume; v, spesific volume; η, viscosity; w, mass fraction; m, molality; x, mole fraction.

The temperature was measured by two TSN-25 platinum resistance thermometers, the pressure by MP-6, MP-60 and MP-600 deadweight pressure gages and by a differential manometer at ambient pressure. In accordance with metrological recommendations of [26], the experimental uncertainties were $\Delta T=\pm3$ mK for temperature, $\Delta p=\pm5\cdot10^{-2}$ MPa for high pressure and $\Delta p=\pm5\cdot10^{-4}$ MPa for ambient pressure, $\Delta\rho=\pm3\cdot10^{-2}$ $kg\cdot m^{-3}$ for density.

The calcium nitrate tetrahydrate (GR grade, >99.0 mass. %) was purchased from Merck (Germany). The solvents were used directly without further purification, whereas the calcium nitrate was desiccated in an oven hold at T=473.15 K under vacuum for the 30 h. Double distilled water was used for the preparation of the solutions. The solutions were prepared by mass using a BP 221 S electronic scale (Sartorius AG, Germany).

3. Results and discussion

In this work, the (p, ρ, T) and (p_s, ρ_s, T_s) properties of $Ca(NO_3)_2$ (aq) at T=(298.15 to 398.15) K and at pressures up to p=60 MPa are reported. The experiments were carried out at molalities m=(0.18848, 0.32075, 0.52994, 1.07546, 2.03143, and 3.28155) $mol\cdot kg^{-1}$ of calcium nitrate. The obtained (p, ρ, T) and (p_s, ρ_s, T_s) values are listed in the Table 2. The presented (p_s, ρ_s, T_s) values were measured in the saturation point at T=398.15 K.

Using a program for standard thermodynamic analysis to describe the (p, ρ, T) and (p_s, ρ_s, T_s) properties of $Ca(NO_3)_2$ (aq), the equation of state [24] was used:

$$p = A\rho^2 + B\rho^8 + C\rho^{12}, \tag{1}$$

where: A, B and C are the coefficients of Eqn. (1) and all are functions of temperature and molality in the following form:

$$A = \sum_{i=1}^{3} T^i \sum_{j=0}^{5} a_{ij} m^j ;\ B = \sum_{i=0}^{2} T^i \sum_{j=0}^{5} b_{ij} m^j ;\ C = \sum_{i=0}^{2} T^i \sum_{j=0}^{5} c_{ij} m^j . \tag{2}$$

The a_{ij}, b_{ij} and c_{ij} are the coefficients of the polynomials and are tabulated in Table 3. The equation of state defined by equations (1) and (2) reproduces our experimental values with a 0.02246 % average deviation.

Figure 1 shows the plot of density ρ of $Ca(NO_3)_2$ (aq) against pressure p at m=0.52994 $mol\cdot kg^{-1}$. Figure 2 shows the plot of density ρ of $Ca(NO_3)_2$ (aq) against pressure p at T=298.15 K. Figure 3 shows the plot of deviations of experimental density $\rho_{exp.}$ from the calculated by equations (1) and (2) density $\rho_{cal.}$ against pressure p.

The values of experimental density are compared with literature values [3, 9, 10, 18, 19, 22]. Figure 4 shows the deviation of density ρ of $Ca(NO_3)_2$ (aq) from literature values against molality m at T=298.15 K and p=0.1 MPa. The comparison of the newly measured and presented values with literature values at the investigated state parameters interval shows good agreement with [3, 9, 10, 18, 19 and 22]. The average deviation of our results with ICT [3] was 0.0429 %. The results of Ewing and Mikovsky [9] were shown the 0.0339 % average deviation. The results of Rodyansky *etc*. [10] are agreed with presented results in 0.2135 %. The comparison of presented results with results of Akhundov *etc*. [18, 19] was shown 0.016 % average deviation. The average deviation of presented results from the results of Vercher *etc*. [22] was 0.07245 %.

The density of investigated solutions at high pressures are available only in [18, 19]. The comparison of the newly measured results at high pressures with these results shows good agreement. The average deviation of both results with presented results at low pressures is ±0.0025 %. The average deviation increase at high pressures ±0.048 %.

TABLE 2: Experimental (p, ρ, T) and (p_s, ρ_s, T_s) values of $Ca(NO_3)_2$ (aq)

p/MPa	ρ/(kg·m^{-3})	p/MPa	ρ/(kg·m^{-3})	p/MPa	ρ/(kg·m^{-3})
m=0.18848 mol·kg^{-1}					
T=298.15 K		T=348.15 K		T=398.15 K	
00.10	1019.61	00.10	996.02	p_s=0.2306	ρ_s=960.37
04.56	1021.64	05.02	998.32	06.25	963.20
09.58	1023.78	10.75	1000.72	10.23	965.07
19.64	1028.07	21.45	1005.21	19.64	969.50
30.45	1032.67	32.86	1009.99	31.12	974.90
40.57	1036.99	39.74	1012.87	40.03	979.09
49.23	1040.68	49.53	1016.97	50.42	983.98
58.79	1044.75	59.61	1021.20	59.45	988.23
T=323.15 K		T=373.15 K			
00.10	1009.68	00.10	979.36		
05.24	1011.97	04.58	981.57		
11.56	1014.58	09.86	983.87		
18.52	1017.46	19.62	988.15		
29.71	1022.09	30.75	993.02		
41.23	1026.85	39.84	997.00		
51.02	1030.90	50.06	1001.48		
59.56	1034.43	58.98	1005.39		
m=0.32075 mol·kg^{-1}					
T=298.15 K		T=348.15 K		T=398.15 K	
00.10	1035.03	00.10	1010.55	p_s=0.2298	ρ_s=974.73
03.75	1036.70	04.99	1012.80	05.75	977.34
12.45	1040.40	10.05	1014.93	09.25	978.99
21.31	1044.18	19.41	1018.88	20.54	984.33
29.64	1047.73	30.45	1023.53	30.06	988.83
41.29	1052.69	40.31	1027.68	39.61	993.34
50.06	1056.43	50.14	1031.82	48.75	997.66
59.12	1060.29	58.65	1035.40	59.63	1002.81
T=323.15 K		T=373.15 K			
00.10	1024.50	00.10	993.72		
05.12	1026.73	05.12	996.18		
09.74	1028.65	10.04	998.34		
19.84	1032.84	21.67	1003.47		
29.95	1037.04	31.62	1007.85		
41.12	1041.69	40.29	1011.67		
50.02	1045.38	51.02	1016.40		
59.83	1049.46	59.61	1020.18		
m=0.52994 mol·kg^{-1}					
T=298.15 K		T=348.15 K		T=398.15 K	
00.10	1058.67	00.10	1032.98	p_s=0.2286	ρ_s=996.86
02.75	1059.87	05.46	1035.52	04.67	998.97
11.45	1063.58	11.94	1038.27	10.63	1001.81
19.56	1067.04	21.54	1042.35	21.37	1006.92
29.34	1071.22	30.08	1045.97	29.64	1010.86
40.27	1075.88	41.36	1050.76	40.37	1015.96
51.64	1080.73	50.34	1054.57	49.61	1020.36
59.03	1083.88	57.96	1057.81	58.37	1024.53

CONTINUE OF TABLE 2

p/MPa	ρ/(kg·m^{-3})	p/MPa	ρ/(kg·m^{-3})	p/MPa	ρ/(kg·m^{-3})
T=323.15 K		T=373.15 K			
00.10	1047.37	00.10	1015.87		
04.96	1049.57	06.87	1019.15		
11.64	1052.37	12.56	1021.68		
20.05	1055.88	19.54	1024.78		
30.12	1060.09	28.73	1028.87		
40.56	1064.46	41.94	1034.74		
50.34	1068.55	50.91	1038.72		
58.43	1071.93	59.76	1042.66		
m=1.07546 mol·kg^{-1}					
T=298.15 K		T=348.15 K		T=398.15 K	
00.10	1116.96	00.10	1088.27	p_s=0.2253	ρ_s=1051.41
03.45	1118.47	04.56	1090.47	05.67	1054.03
11.23	1121.79	09.94	1092.79	10.64	1056.42
19.62	1125.36	19.57	1096.95	20.34	1061.09
29.98	1129.77	29.64	1101.30	30.94	1066.19
40.61	1134.30	41.85	1106.57	40.38	1070.73
50.06	1138.33	52.47	1111.15	50.07	1075.39
59.67	1142.43	59.96	1114.39	59.31	1079.84
T=323.15 K		T=373.15 K			
00.10	1103.90	00.10	1070.57		
05.26	1106.22	05.87	1073.35		
10.34	1108.37	10.67	1075.52		
20.75	1112.76	20.34	1079.88		
30.64	1116.94	30.67	1084.55		
40.08	1120.93	41.31	1089.35		
50.94	1125.52	50.94	1093.70		
59.87	1129.29	59.03	1097.35		
m=2.03143 mol·kg^{-1}					
T=298.15 K		T=348.15 K		T=398.15 K	
00.10	1207.98	00.10	1175.06	p_s=0.2195	ρ_s=1136.71
05.03	1210.00	05.34	1177.53	04.68	1138.87
10.85	1212.44	11.64	1180.28	09.57	1141.24
21.06	1216.71	21.57	1184.62	19.61	1146.11
30.74	1220.75	30.51	1188.53	29.78	1151.04
41.06	1225.07	40.28	1192.79	39.61	1155.81
50.09	1228.84	50.07	1197.07	49.27	1160.49
59.74	1232.88	59.46	1201.17	59.37	1165.39
T=323.15 K		T=373.15 K			
00.10	1192.56	00.10	1156.23		
04.97	1194.70	05.08	1158.72		
09.56	1196.64	11.61	1161.70		
20.37	1201.21	20.07	1165.57		
31.57	1205.94	29.51	1169.88		
40.95	1209.91	40.05	1174.69		
50.27	1213.84	49.96	1179.22		
59.87	1217.90	58.64	1183.18		

p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}	p/MPa	ρ/kg·m^{-3}
m=3.28155 mol·kg^{-1}					
T=298.15 K		T=348.15 K		T=398.15 K	
00.10	1307.58	00.10	1270.97	p_s=0.2109	ρ_s=1230.55
04.96	1309.62	05.02	1273.28	05.06	1233.32
11.37	1312.24	10.34	1275.60	10.27	1235.91
18.56	1315.17	20.17	1279.88	21.34	1241.42
27.54	1318.84	31.20	1284.68	29.58	1245.52
40.31	1324.05	41.06	1288.97	39.57	1250.49
50.62	1328.25	49.21	1292.51	50.67	1256.01
59.12	1331.72	59.31	1296.91	59.11	1260.21
T=323.15 K		T=373.15 K			
00.10	1290.28	00.10	1251.06		
05.03	1292.36	04.64	1253.34		
10.21	1294.49	10.28	1255.94		
20.54	1298.73	20.34	1 260.59		
31.23	1303.11	30.61	1265.33		
40.54	1306.93	39.95	1269.64		
50.24	1310.91	49.27	1273.94		
59.41	1314.67	59.95	1278.87		

TABLE 3: Values of coefficients a_{ij}, b_{ij}, and c_{ij} in the Eqn. (2)

a_{ij}	b_{ij}	c_{ij}
a_{10} = -7.7203424525	b_{00} = 3534.673	c_{00} = -2100.9597
a_{11} = -22.7991105	b_{01} = 7732.784893	c_{01} = -5804.09074161
a_{12} = 70.4184005	b_{02} = -33587.6411462751	c_{02} = 25555.751
a_{13} = -75.13357063	b_{03} = 37840.9591422	c_{03} = -29079.10855
a_{14} = 31.89594533224	b_{04} = -16359.699	c_{04} = 12602.60323706
a_{15} = -4.53723906	b_{05} = 2346.6509433	c_{05} = -1807.877
a_{20} = 0.02717761755	b_{10} = -14.278840747	c_{10} = 9.50409278
a_{21} = 0.0903276715	b_{11} = -36.7610808	c_{11} = 34.7178167
a_{22} = -0.2824457527154	b_{12} = 153.984594	c_{12} = -140.551607431
a_{23} = 0.30305710742	b_{13} = -172.9739773	c_{13} = 157.81198471
a_{24} = -0.129212512	b_{14} = 74.853371	c_{14} = -68.1157257
a_{25} = 0.018439455341	b_{15} = -10.7524183	c_{15} = 9.7561385453
a_{30} = -0.25849·10^{-4}	b_{20} = 0.017998	c_{20} = -0.011613
a_{31} = -0.104342582·10^{-3}	b_{21} = 0.0665418063	c_{21} = -0.06747771
a_{32} = 0.32266154455·10^{-3}	b_{22} = -0.247447162	c_{22} = 0.24359
a_{33} = -0.346607866·10^{-3}	b_{23} = 0.272297762	c_{23} = -0.2676481
a_{34} = 0.14825734275·10^{-3}	b_{24} = -0.1170406841	c_{24} = 0.11464368847
a_{35} = -0.2121646·10^{-4}	b_{25} = 0.01676415	c_{25} = -0.016363424

4. Conclusion

The (p, ρ, T) and (p_s, ρ_s, T_s) properties of $Ca(NO_3)_2$ (aq) at T=(298.15 to 398.15) K, at pressures up to 60 MPa are reported. The experiments were carried out at molalities m=(0.18848, 0.32075, 0.52994, 1.07546, 2.03143, and 3.28155) mol·kg^{-1} of calcium nitrate. The obtained results are compared with corresponding literature values.

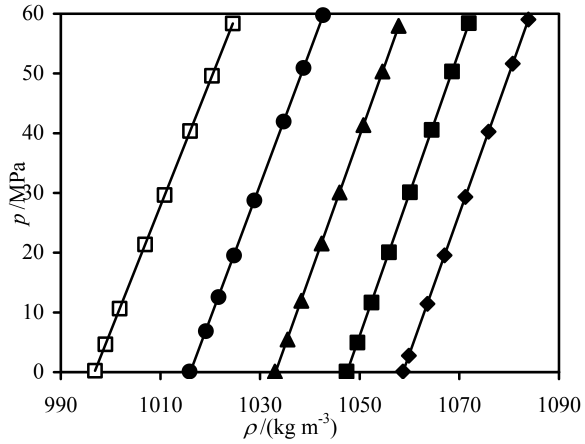

FIGURE 1. Plot of density ρ of $Ca(NO_3)_2$ (aq) against pressure p at m=0.52994 mol·kg^{-1} (◆, 298.15 K; ■, 323.15 K; ▲, 348.15 K; •, 373.15 K; □, 398.15 K).

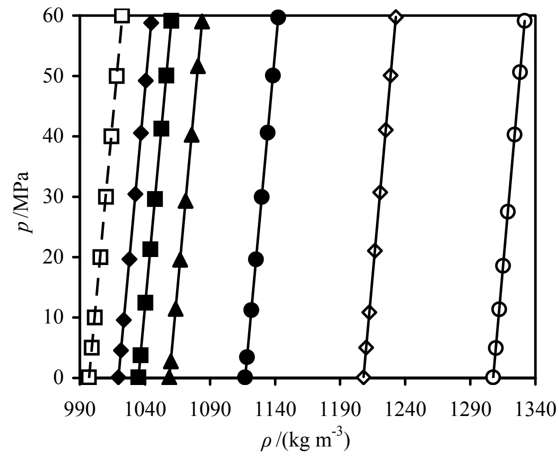

FIGURE 2. Plot of density ρ of $Ca(NO_3)_2$ (aq) against pressure p at T=298.15 K (□, m=0; ◆, m=0.18848 mol·kg^{-1}; ■, m=0.32075 mol·kg^{-1}; ▲, m=0.52994 mol·kg^{-1}; •, m=1.07546 mol·kg^{-1}; ◇, m=2.03143 mol·kg^{-1}; ○, m=3.28155 mol·kg^{-1}).

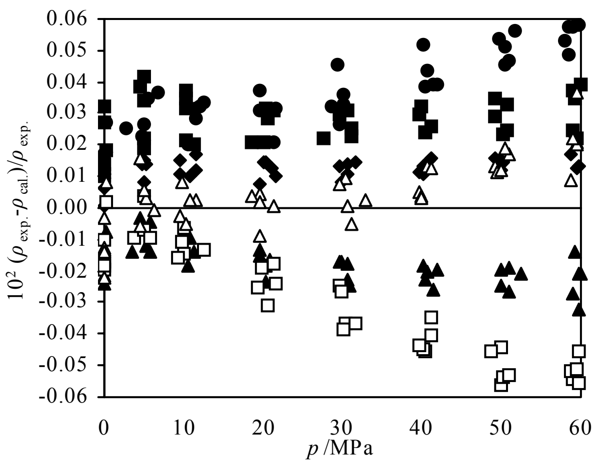

FIGURE 3. Plot of deviations of experimental density $\rho_{exp.}$ from the calculated by equations (1) and (2) density $\rho_{cal.}$ against pressure p (■, m=0.18848 mol·kg^{-1}; ◆, m=0.32075 mol·kg^{-1}; ▲, m=0.52994 mol·kg^{-1}; •, m=1.07546 mol·kg^{-1}; □, m=2.03143 mol·kg^{-1}; △, m=3.28155 mol·kg^{-1}).

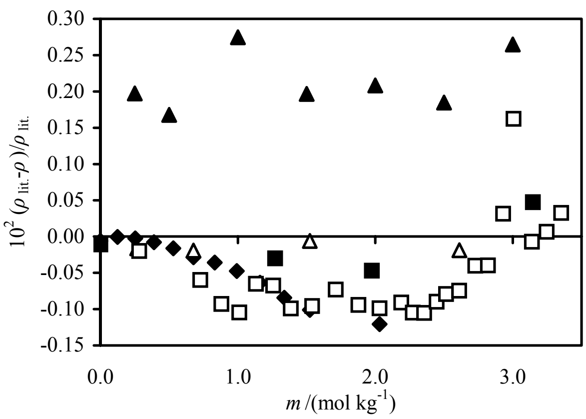

Figure 4. Deviation of density ρ of $Ca(NO_3)_2$ (aq) at T=298.15 K and p=0.1 MPa against molality m, together with values reported in the literature (◆, ICT [5]; ■, Ewing and Mikovsky [9]; ▲, Rodyanskiy *etc.* [10]; △, Akhundov *etc.* [18, 19]; □, Vercher *etc.* [21]).

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$Ca(NO_3)_2$ (sulu) ALTERNATİV ENERJİ MƏNBƏƏLƏRİNDƏ İSTİLİKDAŞIYICILARI KİMİ. I. (p, ρ, T) və (p_S, ρ_S, T_S) XASSƏLƏRİ

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$Ca(NO_3)_2$ (sulu) qarışıqlarının T=(298.15-dən 398.15) K və p=60 MPa təzyiqə qədər (p, ρ, T) və (p_s, ρ_s, T_s) xassələri təqdim olunmuşdur. Təcrübələr m=(0.18848, 0.32075, 0.52994, 1.07546, 2.03143, and 3.28155) mol·kq^{-1} molyar konsentraysiyada aparılmışdır.

$Ca(NO_3)_2$ (вод.) КАК ТЕПЛОНОСИТЕЛЬ ДЛЯ АЛЬТЕРНАТИВНЫХ ИСТОЧНИКОВ ЭНЕРГИИ. I. (p, ρ, T) и (p_S, ρ_S, T_S) СВОЙСТВА

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(p, ρ, T) и (p_s, ρ_s, T_s) свойства $Ca(NO_3)_2$ (вод.) в T=(298.15 до 398.15) K, давлениях до p=60 MPa приведены. Эксперименты проведены в m=(0.18848, 0.32075, 0.52994, 1.07546, 2.03143, и 3.28155) моль · кг$^{-1}$ молярной концентрации кальция нитрата.