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Solubilities of magnesium, calcium, barium, cobalt, nickel, copper, and zinc acetates in water from $T = (278.15 \text{ to } 348.15)$ K

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Solubilities of magnesium acetate, calcium acetate, barium acetate, cobalt acetate, nickel acetate, copper acetate, and zinc acetate in water were determined in the temperature range $T = (278.15 \text{ to } 348.15)$ K and compared with the literature data. © 1999 Academic Press

KEYWORDS: solubilities; metal acetates

1. Introduction

Metal acetates are used in many industrial processes, in preserving food, as mordants for textile printing and dyeing, additives in lubricating oils and greases, catalysts and intermediates in organic reactions, and as pigments for ceramics.(1) Unfortunately, no systematic solubilities of the acetates in water as a function of temperature exist in the main tabulations of solubilities,(2–5) and in some cases the data are even contradictory. On the other hand, solubilities of metal acetates were widely determined, especially by the Stoilova–Balarew group,(6–17) however, only in the context of double salt formation and phase diagrams at fixed temperatures.

Magnesium acetate solubilities were reported in 1926 by Rivett(18) and recently by Apelblat(19) (at constant temperatures(8–10, 15, 20, 21)), but, because the scattering of results is large, the system was reconsidered. The case of calcium acetate was investigated in 1887 by Krasnicki,(22) in 1902 by Lumsden,(23) and new solubilities were presented by Saury *et al*.(24) at fixed temperatures.(10, 12, 16, 20, 25, 26) Solubilities of barium acetate in water were determined in 1887 by Krasnicki,(22) and in 1903 by Walker and Fyffe,(27) and at constant

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temperatures.(14, 15, 20, 28) The Krasicki solubilities are considered by Seidel and Linke(3) as incorrect. Solubilities of cobalt acetate,(6, 7, 11, 13, 14, 17, 29) nickel acetate,(7, 11, 13, 17) copper acetate,(12, 30, 44) and zinc acetate(6–9, 13, 31, 45) are known only at one or two temperatures.

In this work, which is a continuation of our studies of solubilities of organic compounds of biological and industrial importance,(19, 32–36) the solubilities of magnesium, calcium, barium, cobalt, nickel, copper, and zinc acetates are presented.

2. Experimental

Magnesium acetate tetrahydrate, $Mg(CH_3CO_2)_2 \cdot 4H_2O$; barium acetate, $Ba(CH_3CO_2)_2$; cobalt acetate tetrahydrate, $Co(CH_3CO_2)_2 \cdot 4H_2O$; nickel acetate tetrahydrate, $Ni(CH_3CO_2)_2 \cdot 4H_2O$; copper acetate monohydrate, $Cu(CH_3CO_2)_2 \cdot H_2O$; zinc acetate dihydrate, $Zn(CH_3CO_2)_2 \cdot 2H_2O$, all mass fraction >0.99, and calcium acetate $Ca(CH_3CO_2)_2$ mass fraction 0.935 to 0.945 were supplied by Merck and used without further purification.

The solubility measurements were performed with doubly distilled water (in both directions, by increasing and decreasing temperature) as described elsewhere.(32, 36) Weighed samples of saturated solutions of magnesium acetate, calcium acetate, cobalt acetate, and zinc acetate were titrated complexometrically with EDTA (solutions were prepared using $C_{10}H_{14}N_2O_8Na_2 \cdot 2H_2O$). Barium acetate and nickel acetate were determined gravimetrically as $BaSO_4$ and $Ni(C_4H_7O_2N_2)_2$, respectively, and copper acetate by electrolysis. At each temperature, the reported solubilities are the average of two experiments.

3. Results and discussion

The solubilities of metal acetates determined in this work and from the literature(6–31) are presented in table 1 and shown in figures 1 to 7. Both sets of our measured solubilities of magnesium acetate are somewhat lower than those in the literature (figure 1) with the Rivett (1926)(18) values being the highest, differing significantly from results presented here. It is worthwhile to note that in this work the magnesium acetate solubilities are less scattered than we previously reported.(19) Solubilities of magnesium acetate in water as a function of temperature T were correlated by:

$$\begin{aligned} \ln\{(m/m^{\circ}), \mathrm{Mg(CH_3CO_2)_2}\} &= -23.046 + 490.38(T/\mathrm{K})^{-1} + 4.015\ln(T/\mathrm{K}), \\ & 280.15\ \mathrm{K} < T < 348.15\ \mathrm{K}, \end{aligned} \tag{1}$$

where $m^{\circ} = 1\ \mathrm{mol \cdot kg^{-1}}$. The coefficients of this and similar equations were evaluated by an unweighted multivariate least-squares method using only our solubilities from table 1.

The reported solubilities of calcium acetate agree very well with those of previous investigations of Krasnicki (1887)(22) and Lumsden (1902)(23) but surprisingly not with new solubilities of Saury *et al.* (1993)(24) (figure 2). Their statement that “The solubilities and nature of the two former phases were investigated only once in 1902 (*i.e.* by Lumsden) but it appears now that both the solubilities and nature of the solid phase in equilibrium with solution were not properly determined” is clearly incorrect at least with regard to solubilities when the question about the nature of the solid phase is still unsolved. The

TABLE 1. Solubilities m of metal acetates in water as a function of temperature T

T/K	$m/(\mathrm{mol\cdot kg^{-1}})$	T/K	$m/(\mathrm{mol\cdot kg^{-1}})$	T/K	$m/(\mathrm{mol\cdot kg^{-1}})$
		magnesium acetate			
273.15	3.99(18)	295.01	4.21(19)	314.10	4.64(19)
273.15	3.98(21)	298.05	4.61(18)	318.15	5.69(18)
280.15	3.78	298.15	4.61(8)	318.15	5.17
283.15	3.89	298.15	4.40	323.15	5.29
284.25	3.53(19)	300.85	4.20(19)	328.15	6.87(18)
285.52	3.91(19)	303.15	4.59	328.15	5.49
288.05	4.30(18)	304.94	4.24(19)	333.15	5.68
288.15	4.03	308.15	5.04(18)	338.15	5.91
288.22	3.95(19)	308.15	4.79	343.15	6.23
291.88	3.97(19)	308.49	4.54(19)	348.15	6.55
293.15	4.19	311.57	4.58(19)		
293.15	4.45(21)	313.15	4.94		
		calcium acetate			
273.15	2.3646(23)	298.15	2.2329(26)	331.15	1.9916(24)
274.15	2.3938(24)	298.15	2.1738	333.15	2.0674(23)
278.15	2.3172(23)	302.15	2.1607(22)	333.15	1.9220(24)
280.15	2.2893	303.15	2.1382(23)	333.15	2.0704
283.15	2.2657	303.15	2.1420	335.15	1.8904(24)
283.15	2.2748(23)	308.15	2.1180(23)	338.15	2.0725(23)
283.65	2.1876(24)	308.15	2.1227	338.15	2.0696
288.15	2.2331(23)	312.25	2.0295(24)	342.15	1.8841(24)
288.15	2.1243(25)	313.15	2.1003(23)	343.15	2.0851(23)
289.15	2.2350	315.15	2.1076	343.15	2.0648
292.15	2.2000(22)	318.15	2.0886(23)	345.95	1.8588(24)
293.15	2.1958(23)	318.15	2.0886	348.15	2.1003(23)
293.15	2.1971	323.15	2.0750(23)	348.15	2.0614
297.85	2.0232(24)	323.15	2.0762	348.65	1.8588(24)
298.15	2.1623(23)	324.15	2.1343(22)	353.15	2.1180(23)
298.15	2.0918(6)	328.15	2.0674(23)	359.15	1.8335(24)
298.15	2.0906(20)	328.15	2.0738		

Krasnicki and other works(10, 12, 16, 20, 25, 26) were neglected by Saury *et al*. but what is more important is that calcium acetate was determined in old investigations gravimetrically contrary to the EDTA titrations used in Saury *et al*. and in this work. Thus, solubilities of calcium acetate are confirmed by two independent analytical techniques. The form of the

TABLE 1—*continued*

T/K	$m/(\mathrm{mol\cdot kg^{-1}})$	T/K	$m/(\mathrm{mol\cdot kg^{-1}})$	T/K	$m/(\mathrm{mol\cdot kg^{-1}})$
		barium acetate			
273.45	2.3020(27)	298.15	2.8835	328.15	2.9300
273.95	2.2882(22)	299.35	2.9910(27)	333.15	2.9280
278.15	2.3215	299.95	2.8047(22)	319.15	3.0027(22)
280.95	2.4547(22)	300.65	2.8183(22)	322.65	3.0255(22)
281.05	2.4116(27)	303.15	2.9384(15)	323.15	2.9665
283.15	2.5089(22)	303.15	2.9780	324.95	2.9950(27)
283.15	2.4460	303.75	2.9401(27)	336.15	2.9206(27)
288.15	2.5750	308.15	2.9675(27)	337.15	3.0532(22)
290.65	2.7092(27)	308.15	3.0215	338.65	3.0452(22)
293.15	2.8677(14)	312.75	3.0498(27)	346.15	2.8775(27)
293.15	2.7525	313.15	3.0810	351.15	2.9908(22)
294.75	2.8501(22)	313.65	3.0928(27)	353.15	2.9729(22)
297.25	3.0576(27)	314.65	3.0811(27)	357.15	2.8971(27)
298.15	2.9288(20)	317.65	3.0498(27)		
298.15	2.9434(28)	318.15	3.0330		
		cobalt acetate			
278.15	1.2487	298.15	1.5580	333.15	1.8760(13)
283.15	1.3273	303.15	1.7037	333.15	2.1935
288.15	1.3977	308.15	1.8263	338.15	2.2304
293.15	1.3423(14)	313.15	2.0010	343.15	2.2605
293.15	1.4833	318.15	2.1257	348.15	2.4908(29)
298.15	1.4238(6)	323.15	2.1557		
298.15	1.5472(29)	328.15	2.1803		

solubility curve suggests changes in the solid phase and the solubility of calcium acetate in water is expressed for two regions by the equations:

$$\ln\{(m/m^{\mathrm{o}}),\ \mathrm{Ca(CH_3CO_2)_2}\} = -0.019085 + 237.21(T/\mathrm{K})^{-1}, \qquad T < 315\ \mathrm{K}, \quad (2a)$$

$$\ln\{(m/m^{\mathrm{o}}),\ \mathrm{Ca(CH_3CO_2)_2}\} = 0.63077 + 32.301(T/\mathrm{K})^{-1}, \qquad T > 315\ \mathrm{K}. \quad (2b)$$

At room temperature, the composition of the solid phase is not definitively established. The formation of calcium acetate dihydrate, or calcium acetate monohydrate, or calcium acetate hemihydrate, $\mathrm{Ca(CH_3CO_2)_2\cdot 2H_2O}$,(6, 10, 12, 16, 20, 23, 39) $\mathrm{Ca(CH_3CO_2)_2\cdot H_2O}$(11, 20, 22, 24, 26, 38, 40), or $\mathrm{Ca(CH_3CO_2)_2\cdot (1/2)H_2O}$,(20, 24, 26, 41–43), respectively, is postulated based on the solubility, X-ray, and thermal analysis measurements. Lumsden(23) noted that the dihydrate is stable up to $T = 357.15$ K ($T = 356.45$ K is given by Broul *et al.*(39)), calcium acetate monohydrate exists up to $T = 331.15$ K,(24)

TABLE 1—*continued*

T/K	$m/(\mathrm{mol\cdot kg^{-1}})$	T/K	$m/(\mathrm{mol\cdot kg^{-1}})$	T/K	$m/(\mathrm{mol\cdot kg^{-1}})$
nickel acetate					
278.15	0.5860	303.15	0.8315	333.15	1.0775(17)
283.15	0.6300	308.15	0.8935	333.15	1.4485
288.15	0.6955	313.15	0.9186(17)	338.15	1.7105
293.15	0.7372(17)	313.15	0.9385	343.15	1.1833(17)
293.15	0.7340	318.15	1.0290	343.15	1.9850
298.15	0.7465(17)	323.15	0.9795(17)	353.15	1.2798(17)
298.15	0.7965	323.15	1.1475		
303.15	0.8252(17)	328.15	1.2520		
copper acetate					
280.15	0.3292	298.15	0.3842(44)	323.15	0.4775
284.15	0.3373	298.15	0.3771	328.15	0.4906
288.15	0.3455	303.15	0.4055	334.15	0.5669
293.15	0.3595	308.15	0.4175	338.15	0.5964
298.15	0.2837(12)	313.15	0.4360	344.15	0.6497
298.15	0.4010(30)	317.15	0.4542	348.15	0.6912
zinc acetate					
273.15	1.6280(23)	294.15	1.8393	320.15	2.4876
280.15	1.6524	298.15	1.6873(6)	325.15	2.7069
284.15	1.6888	298.15	1.8920	333.15	2.8404(13)
288.15	1.6980(45)	303.15	1.9966	333.15	2.9049
288.15	1.7171	308.15	2.1598	338.15	3.1566
293.15	1.8655(31)	312.15	2.2377	343.15	3.2956
293.15	1.6623(14)	315.15	2.3222	348.15	3.4816

and above $T = 331.15$ K the hemihydrate exists. Panzer(41) postulated that the change from the monohydrate to hemihydrate occurs between $T = 311.15$ K and $T = 339.15$ K. Since the solid phase was not analysed in this work, only the temperature of the change, which is about $T = 315$ K, can be deduced from the solubilities presented here.

Solubilities of barium acetate presented here (figure 3) agree relatively well with the results of Krasnicki (1887),(22) Walker and Fyffe (1903),(27) and with measurements at fixed temperatures.(14, 15, 20, 28) As can be seen in figure 3, the Seidel and Linke(3) remark that the Krasnicki solubilities are incorrect is true only above $T = 335$ K considering that our and the Walker and Fyffe results form a common curve for $T > 315$ K. Barium acetate solubilities as a function of temperature can be expressed by three equations:

$$\ln\{(m/m^{\circ}),\ \mathrm{Ba(CH_3CO_2)_2}\} = 4.1142 - 911.07(T/\mathrm{K})^{-1}, \qquad T < 300.5\ \mathrm{K}, \tag{3a}$$

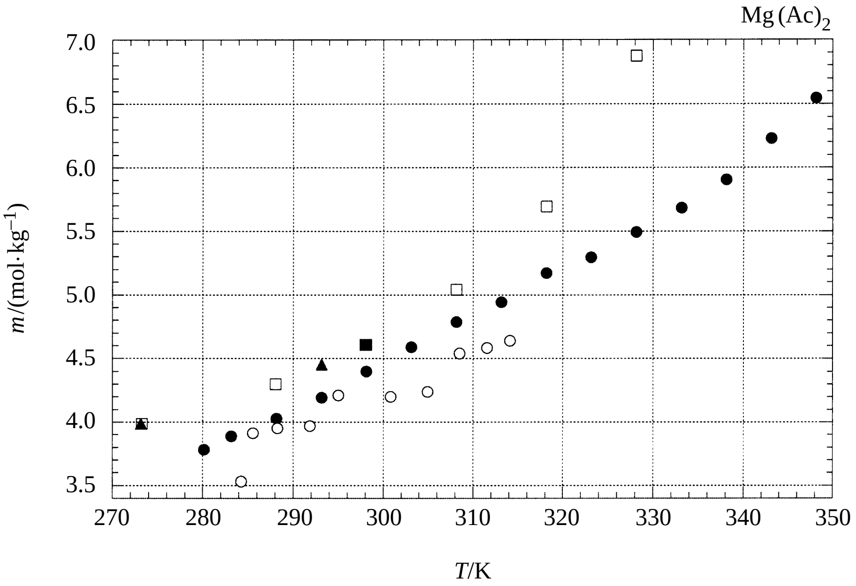

FIGURE 1. Solubility m of magnesium acetate in water as a function of temperature T: ■, reference 8; □, reference 18; ○, reference 19; ▲, reference 21; ●, this work.

$$\ln\{(m/m^{o}), \mathrm{Ba(CH_3CO_2)_2}\} = 2.1132 - 309.81(T/\mathrm{K})^{-1}, \qquad 300.5\ \mathrm{K} < T < 313.4\ \mathrm{K}, \tag{3b}$$

$$\ln\{(m/m^{o}), \mathrm{Ba(CH_3CO_2)_2}\} = -0.004815 + 353.89(T/\mathrm{K})^{-1}, \qquad T > 313.4\ \mathrm{K}. \tag{3c}$$

According to Walker and Fyffe,(27) the solid phase contains barium acetate trihydrate below $T = 297.15$ K (supported also by Stoilova(14) at $T = 293.15$ K), barium acetate dihydrate up to $T = 313.65$ K (observed by Zlatova and Spasova(20) at $T = 298.15$ K), and above this temperature, anhydrous barium acetate exists. Krasnicki(22) mentioned only formation of the dihydrate when Broul *et al.*(39) attributed the change at $T = 297.85$ K from barium acetate trihydrate to barium acetate monohydrate.

The solubility curve of cobalt acetate has two distinct branches (figure 4) defined by:

$$\ln\{(m/m^{o}), \mathrm{Co(CH_3CO_2)_2}\} = -154.572 + 5874.8(T/\mathrm{K})^{-1} + 23.752\ln(T/\mathrm{K}), \qquad 280.15\ \mathrm{K} < T < 317.15\ \mathrm{K}, \tag{4a}$$

and,

$$\ln\{(m/m^{o}), \mathrm{Co(CH_3CO_2)_2}\} = 1.5747 - 260.82(T/\mathrm{K})^{-1}, \qquad 317.15\ \mathrm{K} < T < 343.15\ \mathrm{K}. \tag{4b}$$

Cobalt acetate solubilities in water are known only at few temperatures,(6, 7, 11, 13, 14, 17, 29) and they are, with the exception of the Tudorovskaya *et al.* result(29) at $T = 298.15$ K, systematically lower (especially at higher temperatures) than ours (figure 4). Depressed solubilities of the Stoilova–Balarew group(6–17) result from the fact that acetic acid was

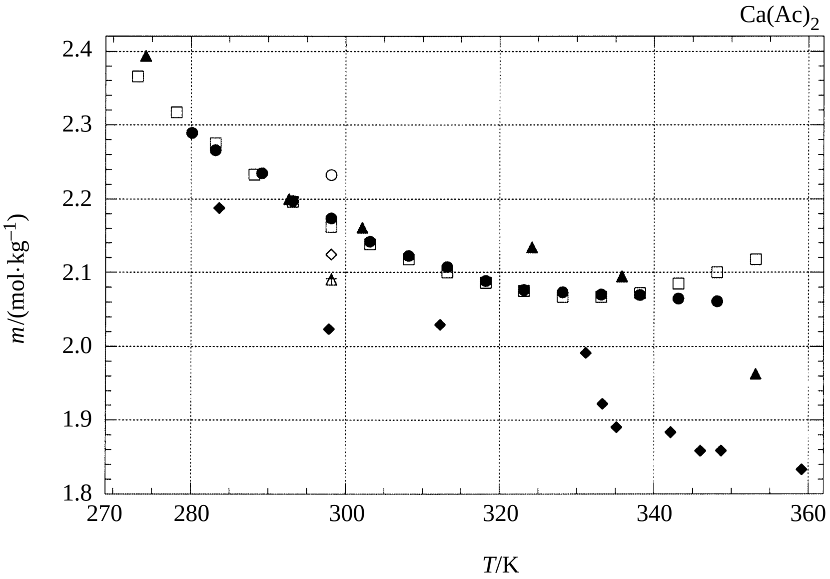

FIGURE 2. Solubility m of calcium acetate in water as a function of temperature T: +, reference 6; △, reference 20; ▲, reference 22; □, reference 23; ◆, reference 24; ◇, reference 25; ○, reference 26; ●, this work.

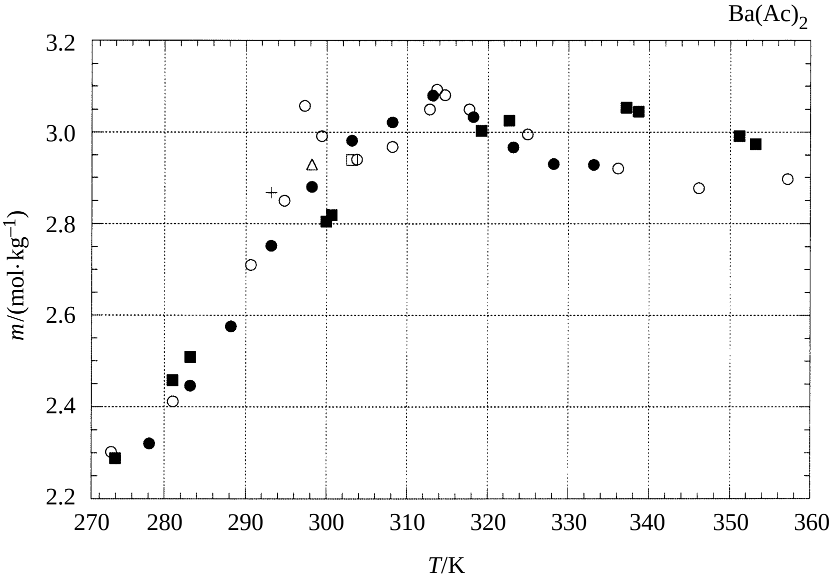

FIGURE 3. Solubility m of barium acetate in water as a function of temperature T: +, reference 14; □, reference 15; △, reference 20; ■, reference 22; ○, reference 27; ●, this work.

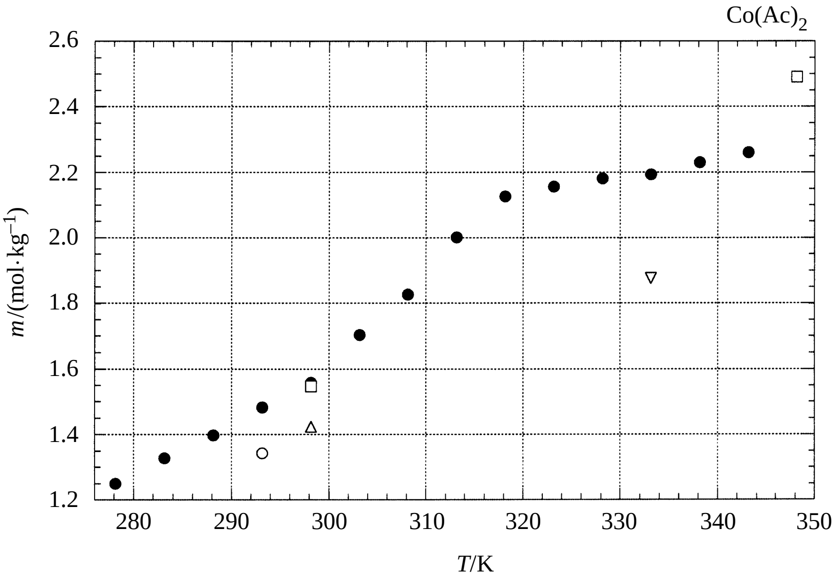

FIGURE 4. Solubility m of cobalt acetate in water as a function of temperature T: △, reference 6; ▽, reference 13; ●, reference 14; □, reference 29; ●, this work.

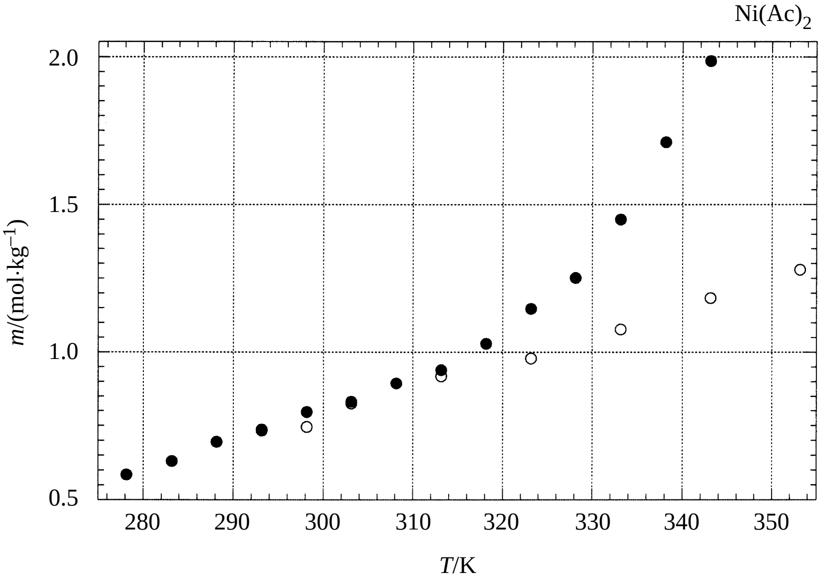

FIGURE 5. Solubility m of nickel acetate in water as a function of temperature T: ○, reference 17; ●, this work.

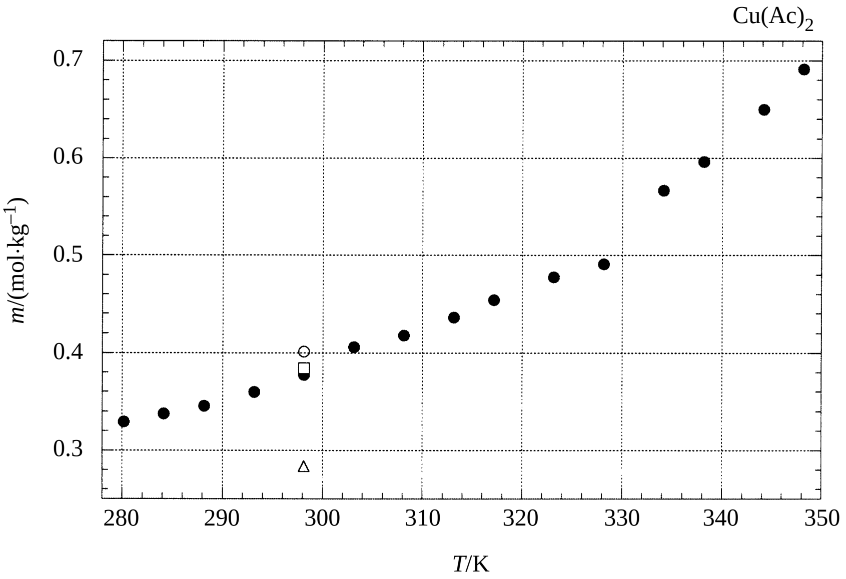

FIGURE 6. Solubility m of copper acetate in water as a function of temperature T: △, reference 12; ○, reference 30; □, reference 44; ●, this work.

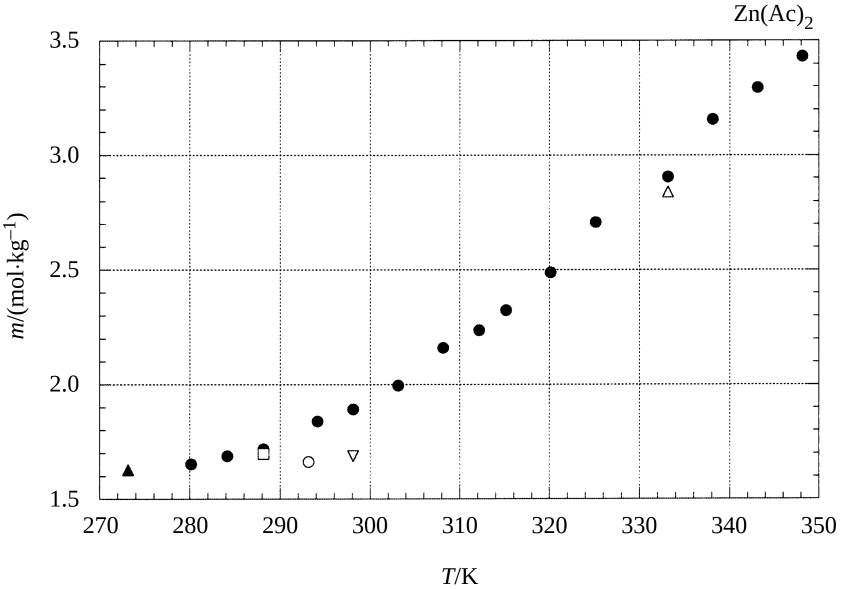

FIGURE 7. Solubility m of zinc acetate in water as a function of temperature T: ▽, reference 6; △, reference 13; ○, reference 14; □, reference 45; ●, this work.

added (about 2 to 4 per cent) in experiments to suppress the possible hydrolysis of metal acetates.

The effect of addition of acetic acid is particularly evident in the case of nickel acetate where the Stoilova *et al.*(38) solubilities are similar to ours up to about $T = 314$ K but considerably lower at higher temperatures (figure 5). It is worthwhile to note that applied analytical methods were different in both works; the complexometric titration was used by Stoilova *et al.*(38) and by us, the gravimetry of which gives all forms of nickel in the solution. Determined solubilities of nickel acetate in water as a function of a temperature are presented as:

$$\ln\{(m/m^{\mathrm{o}}), \mathrm{Ni(CH_3CO_2)_2}\} = 3.7835 - 1199.6(T/\mathrm{K})^{-1}, \qquad 278.15\ \mathrm{K} < T < 323.9\ \mathrm{K}, \tag{5a}$$

and,

$$\ln\{(m/m^{\mathrm{o}}), \mathrm{Ni(CH_3CO_2)_2}\} = 10.848 - 3487.6(T/\mathrm{K})^{-1}, \qquad T > 323.9\ \mathrm{K}. \tag{5b}$$

Previously, the solubility of copper acetate in water was determined only at $T = 298.15$ K.(30, 44) The results of Sandved(30) and Buttgenbach(44) are consistent with our value when, as can be seen in figure 6, the addition of acetic acid (Stoilova and Vassileva(12) study) strongly reduces solubility of copper acetate. The solubility curve of copper acetate is given by:

$$\ln\{(m/m^{\mathrm{o}}), \mathrm{Cu(CH_3CO_2)_2}\} = -136.144 + 5370.8(T/\mathrm{K})^{-1} + 20.561\ln(T/\mathrm{K}), \quad 280.15\ \mathrm{K} < T < 348.15\ \mathrm{K}. \tag{6}$$

Few solubilities of zinc acetate in water are known in the literature(6–9, 13, 31, 45) and they support the solubility curve in spite of the fact that all solubilities are lower than values reported here (figure 7). Determined solubilities of zinc acetate as a function of a temperature can be expressed as:

$$\ln\{(m/m^{\mathrm{o}}), \mathrm{Zn(CH_3CO_2)_2}\} = -87.252 + 3120.21(T/\mathrm{K})^{-1} + 13.592\ln(T/\mathrm{K}), \quad 280.15\ \mathrm{K} < T < 348.15\ \mathrm{K}. \tag{7}$$

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