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Note

THERMODYNAMICS OF THE SILVER—SILVER TUNGSTATE, SILVER—SILVER PHOSPHATE AND SILVER—SILVER ARSENATE ELECTRODES IN AQUEOUS MEDIUM AT DIFFERENT TEMPERATURES

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As part of our programme for the determination of standard potentials of some second-order electrodes of inorganic acids in aqueous, non-aqueous and mixed solvent media [1], we now report the standard potentials of the silver—silver tungstate, silver—silver phosphate and silver—silver arsenate electrodes in aqueous medium.

As in previous communications [1], the cells with liquid junction

$$\mathrm{Ag{-}AgCl/NaCl(c)//Na_2WO_4}(c/2)\mathrm{/Ag_2WO_4{-}Ag} \tag{A}$$

and

$$\mathrm{Ag{-}AgCl/NaCl(s)//Na_3X}(c/3)\mathrm{/Ag_3X{-}Ag} \tag{B}$$

where X is PO_4 or AsO_4, have been studied in water over a range of temperatures.

EXPERIMENTAL

Sodium chloride (B.D.H., AnalaR) was dried above 100°C for 2 h before use. Sodium tungstate (E. Merck), sodium phosphate (E. Merck) and sodium arsenate (B.D.H., AnalaR) were dried at 120°C, and kept in a vacuum desiccator before use.

Silver salts were prepared from silver nitrate (guaranteed reagent) and sodium salts, and the precipitates were repeatedly washed with conductivity water, and were kept in conductivity water in amber-coloured bottles before use.

Preparation of the silver—silver chloride electrode has been described elsewhere [2]. Silver—silver tungstate, silver—silver phosphate and silver—silver arsenate electrodes were prepared by a method similar to that of the silver—silver oxalate, malonate, succinate, adipate or chromate electrodes [1,3] by applying a paste of silver tungstate, phosphate or arsenate to the spongy silver obtained by the thermal decomposition of silver oxide [4]. The electrodes thus prepared were stored in 0.05 M aqueous sodium tungstate, phosphate or arsenate solution for 24 h. Only those electrodes were used which showed a potential difference of 0.1 mV or less on being mutually compared with each other. Stock solutions of sodium chloride, sodium tungstate, sodium phosphate, and sodium arsenate were prepared on a molar basis. Solutions for EMF measurements were prepared from stock solutions by the double dilution method. The cell vessels were of an all-glass type of the design described earlier [1].

Preparation of the cell solutions, setting up of the cells, EMF and conductance measurements were essentially similar to the methods described earlier [1]. All measurements were made in water baths maintained at the appropriate temperature within ±0.1°C. The reproducibility of the EMF measurements was of the order of ±0.2 mV.

RESULTS AND DISCUSSION

A summary of the EMF data at different temperatures for cells (A) and (B) is presented in Tables 1 and 2. The EMF, E, of cell (A) and that of cell (B) are given by expressions (1) and (2), respectively

$$E = E^0_{\text{cell}} + (RT/2F)\ln(2c) + (RT/2F)\ln(y^2_{\text{Cl}} - /y_{\text{WO}_4^{2-}}) + E_j \tag{1}$$

$$E = E^0_{\text{cell}} + (2RT/3F)\ln(3c) + (2RT/3F)\ln(y^3_{\text{Cl}} - /y_{\text{X}^{3-}}) + E_j \tag{2}$$

where the symbols have their usual significance.

The standard molar potentials, E^0_c, of silver—silver tungstate, silver—silver phosphate and silver—silver arsenate electrodes were obtained by the method of extrapolating [1] the auxiliary functions $E^{0'}_c$ given by eqns. (3) and (4), respectively

$$\begin{aligned} E^{0'}_c &= E - \tfrac{1}{2}k\log(2c) - \tfrac{1}{2}k\left\{\frac{4A\sqrt{3c/2}}{1+\sqrt{3c/2}} - \frac{2A\sqrt{c}}{1+\sqrt{c}}\right\} + E^0_{\text{Ag,AgCl}} - E_j \\ &= E^0_{\text{Ag,Ag}_2\text{WO}_4} + bc \end{aligned} \tag{3}$$

TABLE 1

EMF data of cell (A) in water at different temperatures

10^2C (mole dm^{-3})	t (°C)						
	5	10	15	20	25	30	35
0.5	0.1679	0.1620	0.1560	0.1481	0.1439	0.1350	0.1331
1	0.1770	0.1680	0.1610	0.1540	0.1460	0.1401	0.1360
2	0.1820	0.1761	0.1722	0.1630	0.1580	0.1520	0.1430
2.5	0.1840	0.1779	0.1750	0.1659	0.1612	0.1549	0.1459
3.5	0.1882	0.1820	0.1790	0.1725	0.1666	0.1605	0.1515
4.5	0.1920	0.1820	0.1831	0.1760	0.1700	0.1640	0.1550
5	0.1931	0.1872	0.1845	0.1775	0.1710	0.1648	0.1558
5.5	0.1949	0.1885	0.1851	0.1790	0.1720	0.1651	0.1570
6	0.1960	0.1900	0.1850	0.1800	0.1721	0.1651	0.1581
6.5	0.1975	0.1915	0.1865	0.1806	0.1725	0.1655	0.1586
7	0.1990	0.1930	0.1876	0.1807	0.1735	0.1664	0.1592

TABLE 2

EMF data of cell (B) in water at different temperatures

10^2C (mole dm^{-3})	t (°C)						
	5	10	15	20	25	30	35
X = PO_4							
1	0.1990	0.1922	0.1862	0.1802	0.1741	0.1680	0.1620
2	0.2020	0.1960	0.1900	0.1840	0.1780	0.1719	0.1661
3	0.2102	0.2040	0.1971	0.1910	0.1840	0.1781	0.1719
4	0.2131	0.2065	0.1985	0.1925	0.1855	0.1795	0.1735
5	0.2160	0.2102	0.2040	0.1970	0.1910	0.1860	0.1780
6	0.2171	0.2121	0.2061	0.2000	0.1940	0.1872	0.1801
7	0.2200	0.2140	0.2090	0.2041	0.1982	0.1899	0.1834
8	0.2250	0.2190	0.2130	0.2080	0.2020	0.1920	0.1920
9	0.2295	0.2221	0.2161	0.2102	0.2042	0.1980	0.1930
10	0.2310	0.2260	0.2200	0.2141	0.2079	0.2020	0.1959
X = AsO_4							
1	0.1490	0.1420	0.1360	0.1300	0.1241	0.1180	0.1121
2	0.1571	0.1511	0.1451	0.1391	0.1330	0.1269	0.1210
3	0.1589	0.1530	0.1459	0.1410	0.1350	0.1290	0.1230
4	0.1590	0.1549	0.1481	0.1430	0.1369	0.1315	0.1246
5	0.1630	0.1571	0.1500	0.1449	0.1390	0.1335	0.1265
7	0.1695	0.1615	0.1552	0.1490	0.1425	0.1360	0.1300
8	0.1697	0.1635	0.1370	0.1525	0.1445	0.1380	0.1320
9	0.1710	0.1650	0.1585	0.1532	0.1465	0.1405	0.1345
11	0.1715	0.1670	0.1600	0.1550	0.1486	0.1426	0.1370
14	0.1765	0.1705	0.1650	0.1600	0.1548	0.1480	0.1435
15	0.1771	0.1710	0.1661	0.1610	0.1555	0.1491	0.1440

and

$$E_c^{0'} = E - \tfrac{2}{3}k\log(3c) - k\left\{\frac{3A\sqrt{2c}}{1+\sqrt{2c}} - \frac{A\sqrt{c}}{1+\sqrt{c}}\right\} + E^0_{\mathrm{Ag,AgCl}} - E_j$$

$$= E^0_{\mathrm{Ag,Ag_3X}} + b'c \qquad (4)$$

to the molarity $c = 0$, where $E^0_{\mathrm{Ag,AgCl}}$ is the standard molar potential of the silver—silver chloride electrode and A is the Debye—Huckel constant, and are known [5] over the temperature range under investigation. In eqns. (3) and (4), $k = 2.3026\ (RT/F)$, $b = \frac{1}{2}k\{(2\beta_{\mathrm{Cl^-}}) - (3/2\beta_{\mathrm{WO_4^{2-}}})\}$, and $b' = k\{(\beta_{\mathrm{Cl^-}}) - (2/3\beta_{\mathrm{X^{3-}}})\}$, where β is the usual constant in the Debye—Huckel expression for the activity coefficient, i.e.

$$-\log y_i = \frac{AZ_i^2\mu^{1/2}}{1+\mu^{1/2}} - \beta_i\mu$$

where "i" is Cl^-, WO_4^{2-} or X^{3-}.

As usual [1], the values of the liquid junction potentials, E_j, were calculated from the equivalent conductances of sodium tungstate, sodium phosphate, or arsenate and sodium chloride by means of Lewis and Sargent's equation. These values are given in Table 3.

TABLE 3

Conductance data for NaCl[a], $Na_2WO_4^b$ *, $Na_3PO_4^c$ * and $Na_3AsO_4^d$ * in aqueous medium at different temperatures

10^2C (mole dm^{-3})	10^3 conductance (mho)											
	t (°C)											
	5				10				15			
	a	b	c	d	a	b	c	d	a	b	c	d
0.5	0.8	0.7			0.85	0.75			0.9	0.85		
1	1.15	1.1	0.595	0.7	1.25	1.15	0.65	0.75	1.45	1.5	0.75	0.85
2	1.8	1.9	1.075	1.0	2.15	2.0	1.1	1.1	2.7	2.15	1.2	1.2
2.5	2.2	2.3			2.6	2.425			3.3	2.6		
3	2.5		1.55	1.3	3.1		1.55	1.4	3.9		1.7	1.6
3.5	2.9	3.05			3.5	3.25			4.5	3.5		
4	3.25		1.95	1.55	4.0		2.0	1.75	5.15		2.2	2.0
4.5	3.8	3.85			4.5	4.05			5.8	4.4		
5	4.4	4.25	2.35	1.85	5.0	4.5	2.4	2.1	6.45	4.8	2.68	2.35
5.5	5.05	4.65			5.65	4.9			7.0	5.25		
6	5.65	5.05	2.65		6.35	5.3	2.75		7.67	5.65	3.05	
6.5	6.3	5.45			7.0	5.7			8.3	6.1		
7	6.95	5.85	2.95	2.45	7.75	6.1	3.1	2.85	9.0	6.5	3.45	3.15
8	8.15		3.25	2.75	9.2		3.5	3.2	10.3		3.85	3.55
9	9.45		3.55	3.05	10.5		3.85	3.55	11.5		4.3	3.9
10	10.7		3.9		11.8		4.25		12.8		4.65	
11	12.0			3.65	13.0			4.3	14.0			4.7
14	15.7			4.5	16.9			5.4	17.8			5.85
15	17.0			4.8	18			5.75	19.1			6.25

	20				25				30				35			
	a	b	c	d	a	b	c	d	a	b	c	d	a	b	c	d
0.5	0.95	1.0			1.1	1.1			1.25	1.5			1.3	1.425		
1	1.55	1.5	0.85	1.05	1.8	1.625	1.05	1.1	2.0	1.85	1.25	1.2	2.1	2.05	1.35	1.35
2	2.95	2.5	1.4	1.45	3.3	2.7	1.6	1.55	3.55	3.0	1.8	1.75	3.95	3.35	2.05	1.95
2.5	3.6	3.0			4.0	3.2			4.35	3.6			4.75	4.0		
3	4.3		1.95	1.85	4.7		2.15	2.0	5.1		2.4	2.3	5.6		2.7	2.5
3.5	5.0	3.9			5.5	4.25			6.0	4.75			6.5	5.25		
4	5.7		2.45	2.25	6.3		2.7	2.25	6.8		3.0	2.85	7.4		3.35	3.05
4.5	6.4	4.875			7.0	5.3			7.5	5.9			8.3	6.55		
5	7.1	5.375	2.9	2.7	7.8	5.8	3.25	2.95	8.4	6.5	3.6	3.4	9.15	7.2	3.95	3.65
5.5	7.8	5.85			8.5	6.3			9.2	7.05			10.0	7.85		
6	8.5	6.35	3.45		9.3	6.85	3.8		10.0	7.65	4.25		10.95	8.5	4.65	
6.5	9.2	6.85			10.0	7.35			10.8	8.2			11.8	9.15		
7	9.95	7.3	3.9	3.5	10.8	7.85	4.35	3.9	11.6	8.7	4.9	4.45	12.7	9.8	5.35	4.8
8	11.35		4.35	3.9	12.35		4.9	4.4	13.3		5.4	5.0	14.5		5.95	5.35
9	12.8		4.85	4.35	13.8		5.4	4.85	15.0		6.0	5.5	16.3		6.45	5.95
10	14.3		5.3		15.35		5.85		16.65		6.5		18.1		7.1	
11	15.75			5.15	16.9			5.85	18.3			6.55	19.9			7.05
14	20.25			6.4	21.4			7.25	23.25			8.05	25.3			8.75
15	21.75			6.85	22.9			7.7	24.8			8.6	27.1			9.3

* b, *c*/2; c and d, *c*/3.

TABLE 4

Standard molar potentials (E^0_c in abs. volts) for the $Ag(s)/Ag_2WO_4(s)$, WO_4^{2-}; $Ag(s)/Ag_3PO_4(s)$, PO_4^{3-} and $Ag(s)/Ag_3AsO_4(s)$, AsO_4^{3-} electrodes in water from 5 to 35°C

Electrode	t (°C)						
	5	10	15	20	25	30	35
Silver—silver tungstate	0.4692	0.4598	0.4528	0.4435	0.4351	0.4282	0.4200
Silver—silver phosphate	0.4517	0.4797	0.4675	0.4605	0.4523	0.4456	0.4370
Silver—silver arsenate	0.4391	0.4288	0.4173	0.4114	0.4008	0.3938	0.3849

The values of E^0_c of the silver—silver tungstate, silver—silver phosphate and silver—silver arsenate electrodes, thus obtained are listed in Table 4. The values of the standard deviations calculated by the least-squares method are within ±0.3—0.8 mV. The values for the standard potential on the molal (E^0_m) and mole fraction (E^0_N) scales have also been calculated at experimental temperatures by the usual equations [6].

$$E^0_m = E^0_c - 2(2.3026RT/F) \log \rho$$

and

$$E^0_N = E^0_m - 2(2.3026RT/F) \log(1000/M) \tag{5}$$

where ρ and m are the density and molecular weight, respectively of water. The variation of the standard potential with temperature in molar (E^0_c), molal (E^0_m) and mole fraction (E^0_N) scales has been expressed by equations of

TABLE 5

Constants of eqn. (6) for molar (c), molal (m) and mole fraction (N) scales in water

electrode	X	A	10^2B	-10^2C	10^4D
Silver—silver tungstate	c	1.1760	−0.062505	0.058581	0.099182
	m	1.1378	0.14016	0.096556	0.11700
	N	1.2016	−0.14236	0.060278	0.10705
Silver—silver phosphate	c	2.4068	−0.22377	0.19833	0.46821
	m	2.4550	−0.35939	0.17661	0.46532
	N	2.3890	−0.17280	0.22164	0.48065
Silver—silver arsenate	c	1.6254	−0.24932	0.085987	0.22040
	m	1.5930	−0.069794	0.11986	0.23677
	N	1.6217	−0.24959	0.10038	0.23003

TABLE 6

Values of the standard potentials of silver—silver tungstate, silver—silver phosphate and silver—silver arsenate electrodes on the molar, molal and mole fraction scales in water and thermodynamic quantities on the molal and mole fraction scales at 5, 15, 25 and 35°C

	t (°C)			
	5	15	25	35
Silver—silver tungstate electrode				
E_c^0(V)	0.4692	0.4528	0.4351	0.4200
E_m^0(V)	0.4692	0.4529	0.4352	0.4203
E_N^0(V)	0.2767	0.2534	0.2288	0.2070
$-\Delta G_m^0$(J mole^{-1})	905 15.0	87 227.0	84 100.0	81 137.0
$-\Delta G_N^0$(J mole^{-1})	53 363.0	48 732.9	44 267.0	39 969.0
$-\Delta S_m^0$(J mole^{-1} K^{-1})	336.78	320.78	304.6	288.13
$-\Delta S_N^0$(J mole^{-1} K^{-1})	471.40	454.85	438.16	421.34
$-\Delta H_m^0$(J mole^{-1})	184 140.0	179 610.0	174 860.0	169 880.0
$-\Delta H_N^0$(J mole^{-1})	184 410.0	179 730.0	174 840.0	169 740.0
Silver-silver phosphate electrode				
E_c^0(V)	0.4917	0.4675	0.4523	0.4370
E_m^0(V)	0.4917	0.4675	0.4525	0.4373
E_N^0(V)	0.2992	0.2681	0.2461	0.2240
$-\Delta G_m^0$(J mole^{-1})	142 140.0	135 890.0	130 800.0	126 880.0
$-\Delta G_N^0$(J mole^{-1})	86 420.0	78 157.0	71 063.0	65 145.0
$-\Delta S_m^0$(J mole^{-1} K^{-1})	684.02	567.38	450.13	332.30
$-\Delta S_N^0$(J mole^{-1} K^{-1})	884.44	767.97	650.73	532.76
$-\Delta H_m^0$(J mole^{-1})	332 300.0	299 290.0	264 940.0	229 230.0
$-\Delta H_N^0$(J mole^{-1})	332 290.0	299 330.0	264 980.0	229 230.0
Silver—silver arsenate electrode				
E_c^0(V)	0.4391	0.4173	0.4008	0.3849
E_m^0(V)	0.4391	0.4173	0.4010	0.3852
E_N^0(V)	0.2466	0.2179	0.1946	0.1719
$-\Delta G_m^0$(J mole^{-1})	127 000.0	121 320.0	116 200	111 660.0
$-\Delta G_N^0$(J mole^{-1})	71 278.0	63 587.0	56 461	49 903.0
$-\Delta S_m^0$(J mole^{-1} K^{-1})	596.25	539.97	483.27	426.18
$-\Delta S_N^0$(J mole^{-1} K^{-1})	797.25	740.92	684.25	627.25
$-\Delta H_m^0$(J mole^{-1})	292 760.0	276 830.0	260 220.0	242 920.0
$-\Delta H_N^0$(J mole^{-1})	292 910.0	276 970.0	260 360.0	243 090.0

the type [7]

$$E_x^0 = A + BT + CT\ln T + DT^2/2 \qquad (6)$$

where x is c, m or N, and T is the temperature in Kelvin. The parameters A, B, C, and D of eqn. (6) on different scales are presented in Table 5.

The thermodynamic quantities, ΔG^0, ΔH^0, and ΔS^0 for the electrode reactions

$$Ag_2WO_4(s) + 2\,e \rightleftharpoons 2\,Ag(s) + WO_4^{2-}\ (solvated)$$

and

$$Ag_3X(s) + 3\,e \rightleftharpoons 3\,Ag(s) + X^{3-}\ (solvated) \qquad (7)$$

have been calculated by the usual relations [7], and are given in Table 6 on the molality and mole fraction scales at four different temperatures.

The thermodynamic solubility products, K_{so}^0, of silver tungstate, silver phosphate and silver arsenate in aqueous medium have been calculated from the values of standard potentials by means of the equations

$$\ln K_{so}^0 = [E^0(Ag/Ag_2WO_4) - E^0(Ag/Ag^+)]\,(2F/RT) \qquad (8a)$$

and

$$\ln K_{so}^0 = [E^0(Ag/Ag_3X) - E^0(Ag/Ag^+)]\,(3F/RT) \qquad (8b)$$

where $E^0(Ag/Ag^+)$ is the standard molal potential of the silver electrode and is obtained from the literature [5] over the temperature range under investigation. The values for K_{so}^0 on the molarity (K_{so}^c) and molality (K_{so}^m) scales at the experimental temperatures are shown in Table 7. The data for K_{so}^c were calculated from $K_{so}^m X \rho^2$. The solubility product data were fitted by least squares to the following equations [8]

$$\log K_{so}^c(Ag_2WO_4) = -1742/(T/K) - 6.440 \qquad \sigma = 0.0084 \qquad (9a)$$

TABLE 7

Solubility product constants, on the molar (K_s^c) and molal (K_s^m) scales of Ag_2WO_4, Ag_3PO_4 and Ag_3AsO_4 in water at different temperatures

Silver salt	t (°C)						
	5	10	15	20	25	30	35
Tungstate							
$K_s^c \times 10^{13}$	2.089	2.399	3.375	3.882	4.838	6.604	8.213
$K_s^m \times 10^{13}$	2.09	2.40	3.38	3.90	4.839	6.71	8.31
Phosphate							
$K_s^c \times 10^{18}$	1.333	1.363	1.441	1.719	2.317	3.464	5.111
$K_s^m \times 10^{18}$	1.333	1.367	1.443	1.725	2.331	3.494	5.172
Arsenate							
$K_s^c \times 10^{21}$	2.207	2.599	2.693	5.353	6.149	10.370	14.200
$K_s^m \times 10^{21}$	2.207	2.601	2.695	5.371	6.186	10.580	14.360

TABLE 8

Values of the standard molal potentials, standard thermodynamic quantities and solubility product constants at 25°C

	Silver—silver chromate	Silver—silver tungstate	Silver—silver phosphate	Silver—silver arsenate
E^0_m(V)	0.4491	0.4352	0.4525	0.4010
$-\Delta G^0$(J mole^{-1})	86 690	84 100.0	130 800.0	116 200.0
$-\Delta H^0$(J mole^{-1})	151 960	174 860.0	264 940.0	260 220.0
$-\Delta S^0$(J mole^{-1} K^{-1})	219	304.6	450.13	483.27
K^m_s(mole kg^{-2})	$\frac{Ag_2CrO_4}{2.71 \times 10^{-12}}$	$\frac{Ag_2WO_4}{4.83 \times 10^{-13}}$	$\frac{Ag_3PO_4}{2.33 \times 10^{-18}}$	$\frac{Ag_3AsO_4}{6.19 \times 10^{-21}}$

$$\log K^{m}_{so}(Ag_2WO_4) = -1748/(T/K) - 6.420 \qquad \sigma = 0.00015 \tag{9b}$$

$$\log K^{c}_{so}(Ag_3PO_4) = -1766/(T/K) - 11.6391 \qquad \sigma = 0.0104 \tag{9c}$$

$$\log K^{m}_{so}(Ag_3PO_4) = -1748/(T/K) - 11.6886 \qquad \sigma = 0.00026 \tag{9d}$$

$$\log K^{c}_{so}(Ag_3AsO_4) = -2381/(T/K) - 12.1631 \qquad \sigma = 0.0107 \tag{9e}$$

$$\log K^{m}_{so}(Ag_3AsO_4) = -2365/(T/K) - 12.2206 \qquad \sigma = 0.019 \tag{9f}$$

where σ is the standard deviation for the precision of the least squares fit: the correlation coefficient for these relations is within 0.640—1.384.

Table 8 includes the standard molal potentials and standard thermodynamic quantities of the electrode reactions at 25° C along with those of the silver—silver chromate electrode [1] for the sake of comparison. The solubility product constants of silver chromate [9], silver tungstate, silver phosphate and silver arsenate at 25° C are also included in Table 8. The sequence $E^0_{Ag,Ag_2CrO_4} > E^0_{Ag,Ag_2WO_4}$ and $E^0_{Ag,Ag_3PO_4} > E^0_{Ag,Ag_3AsO_4}$ shows that CrO_4^{2-} and PO_4^{3-} ions are more strongly solvated than their counterparts. The relative solubilities of silver salts containing different anions can also be interpreted on the basis of difference in anionic solvation [10]. The order in the K^0_{so} values, $Ag_2CrO_4 > Ag_2WO_4 > Ag_3PO_4 > Ag_3AsO_4$, would be in accordance with the supposition that the solubility decreases with increase in the size of the anion: larger anions are less solvated.

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