Registry No. Butan-1-ol, 71-36-3; propan-1-ol, 71-23-8; decane, 124-18-5.

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Densities of Aqueous Lithium, Sodium, and Potassium Hydroxides from 25 to 75 °C at 1 atm

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A weight dilatometer has been used to measure the densities of aqueous solutions of lithium, sodium, and potassium hydroxides up to 3 *m* at 10 °C intervals in the range 25–75 °C at 1.013 bar. The precision of the densities is ±1 × 10^{-5} g cm^{-3}. The data are compared with literature values where applicable.

Introduction

In the conversion of fossil and nuclear energy to electricity high-temperature solution-phase thermodynamic data are required to improve plant reliability. For example, most corrosion problems in boiler water circuits arise where the solution can concentrate; the compositions of these concentrated solutions can only be obtained from thermochemical data. In order to obtain density data at high temperatures it is necessary to have a good knowledge of the density at lower temperatures. Currently there is a great lack of solution density data on alkalis at temperatures other than room temperature. In this work we have been interested in developing a technique which can be used to obtain data at atmospheric pressure and that can be relatively easily adapted to measuring densities under pressure at high temperatures. This technique is used to provide the basic low-temperature data for the high-temperature study.

Experimental Section

Materials. The alkali hydroxides used were AR grade. Carbonate-free solutions were prepared by precipitation of carbonate using the method described by Vögel (*1*). The carbonate-free hydroxide solutions were then stored under nitrogen in blacked-out plastic containers until use. The potassium hydrogen phthalate used for standardization was recrystallized 3 times from conductivity water after a hot filtration; it was then dried in the vacuum oven and stored under vacuum until use. All solutions were prepared by using demineralized water which was first distilled and then passed through a nuclear-grade mixed bed in-exchange resin, followed by a filter column until its conductivity was less than 0.05 μS cm^{-1}. All mercury used was recovered and cleaned by a four-stage process, dried by passing air through it in contact with AR grade acetone, and stored under nitrogen in plastic containers until use.

All solutions were degassed by boiling under reduced pressure for $^1/_2$ h before standardization. The hydroxide solutions were standarized by titration against weighed portions of a potassium hydrogen phthalate solution, which had been made up by weight, using a Metrohm 636 Titroprocessor in conjunction with a Metrohm E635 Dosimat unit. This procedure gave a reproducibility of better than 0.005%.

Table I. Density of Water

T/K	ρ_0/g cm^{-3}	T/K	ρ_0/g cm^{-3}
298.15	0.997048	328.15	0.985696
308.15	0.994035	338.15	0.980553
318.15	0.990216	348.15	0.974844

Solutions were made up by weight and buoyancy corrections applied; the air density at the time of weighing was calculated from tables (*2*). Two balances were used during this work: first, a Stanton SM12 double pan balance, used in conjunction with a set of National Physical Laboratory (NPL) calibration stainless steel weights, and second, a Sartorius 2004 MP electronic balance. Both balances gave a precision of ±2 × 10^{-5} g and operated over a range of 0–160 g. The two balances agreed to within ±1 × 10^{-5} g on a given weight.

Dilatometer. The dilatometer used was similar in design to the weight dilatometer of Gibson and Loeffler (*3*), but was made of titanium. The opening for filling the dilatometer was made as narrow as possible and the flange was sealed by a flat plate clamped against a silicone rubber seating. The principle of operation is that mercury is diplaced through a capillary from the dilatometer and collected in a preweighed pot. The advantage of this method is that it allows a large number of measurements to be made over several temperatures without refilling the dilatometer, to give a precision of ±1 × 10^{-5} g cm^{-3}. Values for the density of water were taken from Kell (*4*) (Table I) and for the density of mercury from ref *2*. The temperature of the water thermostat was monitored with an NPL calibrated platinum resistance thermometer, in conjunction with an Automatic Systems Laboratories precision ac double transformer ratio bridge (Model H6), to ±1 × 10^{-3} K. Temperature control was better than ±2 × 10^{-3} K.

Results and Discussion

Values for the density of each electrolyte were obtained at 10-deg intervals between 25 and 75 °C, and over a 0.5–3 *m* concentration range. Measurements were not carried out below 0.5 *m* as a precision of ±1 × 10^{-5} g cm^{-3} in the density gives an error of ±0.02 cm^3 mol^{-1} in the apparent molar volume at 0.5 *m* but ±0.1 cm^3 mol^{-1} at 0.1 *m*. The results for lithium hydroxide, sodium hydroxide, and potassium hydroxide are given in Tables II–IV. The apparent molar volume of each solution was calculated from these data by using eq 1 (see Glossary at end of paper).

$$^{\phi}V = \frac{1}{m}\left(\frac{1}{\rho} - \frac{1}{\rho_0}\right) + \frac{M_2}{\rho} \qquad (1)$$

0021-9568/86/1731-0031$01.50/0

Table II. Densities of Lithium Hydroxide Solutions

	ρ/g cm^{-3}					
m/mol kg^{-1}	298.15 K	308.15 K	318.15 K	328.15 K	338.15 K	348.15 K
0.4800	1.010 05	1.006 88	1.003 00	0.998 48	0.993 38	0.987 77
0.7014	1.015 87	1.012 63	1.008 72	1.004 20	0.999 12	0.993 54
0.9356	1.021 91	1.018 60	1.014 66	1.010 13	1.005 07	0.999 52
1.1314	1.026 87	1.023 50	1.019 53	1.015 00	1.009 95	1.004 43
1.3306	1.031 83	1.028 40	1.024 40	1.019 87	1.014 83	1.009 33
1.3829	1.033 11	1.029 67	1.025 67	1.021 13	1.016 09	1.010 60
1.6851	1.040 43	1.036 91	1.032 86	1.028 31	1.023 28	1.017 81
1.8532	1.044 42	1.040 85	1.036 78	1.032 21	1.027 18	1.021 74
2.2556	1.053 71	1.050 04	1.045 90	1.041 31	1.036 28	1.030 86
2.2587	1.053 78	1.050 11	1.045 97	1.041 38	1.036 35	1.030 93
2.5651	1.060 61	1.056 87	1.052 69	1.048 06	1.043 02	1.037 62
2.7892	1.065 47	1.061 68	1.057 47	1.052 82	1.047 77	1.042 37

Table III. Densities of Sodium Hydroxide Solutions

	ρ/g cm^{-3}					
m/mol kg^{-1}	298.15 K	308.15 K	318.15 K	328.15 K	338.15 K	348.15 K
0.5038	1.018 81	1.015 36	1.011 17	1.006 45	1.001 18	0.995 41
0.7483	1.029 02	1.025 38	1.021 05	1.016 22	1.010 90	1.005 09
0.9999	1.039 30	1.035 48	1.031 03	1.026 10	1.020 72	1.014 88
1.2447	1.049 11	1.045 11	1.040 56	1.035 54	1.030 12	1.024 24
1.3534	1.053 40	1.049 33	1.044 74	1.039 68	1.034 23	1.028 34
1.7039	1.067 01	1.062 72	1.058 02	1.052 83	1.047 32	1.041 38
2.0006	1.078 28	1.073 80	1.069 01	1.063 73	1.058 17	1.052 18
2.2447	1.087 39	1.082 76	1.077 90	1.072 55	1.066 94	1.060 92
2.4929	1.096 51	1.091 74	1.086 80	1.081 40	1.075 77	1.069 69
2.7396	1.105 47	1.100 54	1.095 52	1.090 08	1.084 41	1.078 28
3.0094	1.115 14	1.110 05	1.104 91	1.099 45	1.093 76	1.087 55

Table IV. Densities of Potassium Hydroxide Solutions

	ρ/g cm^{-3}					
m/mol kg^{-1}	298.15 K	308.15 K	318.15 K	328.15 K	338.15 K	348.15 K
0.4920	1.021 28	1.017 87	1.013 78	1.009 10	1.003 82	0.998 04
0.8058	1.036 18	1.032 55	1.028 32	1.023 53	1.018 20	1.012 39
0.9554	1.043 13	1.039 41	1.035 11	1.030 28	1.024 93	1.019 11
1.1128	1.050 36	1.046 54	1.042 18	1.037 29	1.031 92	1.026 11
1.3484	1.060 98	1.057 04	1.052 59	1.047 63	1.042 24	1.036 42
1.7145	1.077 09	1.072 96	1.068 40	1.063 34	1.057 90	1.052 10
1.7830	1.080 05	1.075 89	1.071 31	1.066 23	1.060 78	1.054 98
2.0344	1.090 78	1.086 50	1.081 85	1.076 72	1.071 24	1.065 45
2.3453	1.103 76	1.099 35	1.094 63	1.089 44	1.083 91	1.078 12
2.5502	1.112 16	1.107 65	1.102 90	1.097 69	1.092 11	1.086 31
3.0211	1.131 00	1.126 29	1.121 46	1.116 22	1.110 51	1.104 68

Table V. Fitting Parameters for Lithium Hydroxide

T/K	${}^{\phi}V^{\infty}$/cm^3 mol^{-1}	$10^5(\partial\beta^{\circ}/\partial P)$/ kg mol^{-1} atm^{-1}	$10^5(\partial\beta^1/\partial P)$/ kg^2 mol^{-2} atm^{-1}	$10^6(\partial C^{\phi}/\partial P)$/ kg^2 mol^{-2} atm^{-1}	std dev of fit
298.15	−4.287 ± 0.012	−0.111 ± 0.091	3.759 ± 0.251	−0.335 ± 0.185	0.002
308.15	−4.103 ± 0.008	−0.384 ± 0.057	3.986 ± 0.159	−0.253 ± 0.117	0.001
318.15	−4.128 ± 0.010	−0.760 ± 0.072	4.670 ± 0.200	−0.477 ± 0.147	0.002
328.15	−4.327 ± 0.010	−1.052 ± 0.072	5.109 ± 0.201	−0.313 ± 0.148	0.002
338.15	−4.619 ± 0.013	−1.615 ± 0.085	6.283 ± 0.236	−0.584 ± 0.174	0.002
348.15	−5.168 ± 0.013	−1.469 ± 0.084	5.748 ± 0.232	−0.098 ± 0.171	0.002

The apparent molar volume data were smoothed by fitting to an extended form of the Debye–Hückel equation derived by Pitzer (*5*):

$$
{}^{\phi}V - [{}^{\phi}V^{\text{st}} - {}^{\phi}V^{\infty}] = {}^{\phi}V^{\infty} + 2RTm\left(\frac{\partial\beta^0}{\partial P}\right)_T + \frac{4RTm}{\alpha^2}\left(\frac{\partial\beta^1}{\partial P}\right)_T[1 - (1 + \alpha m^{1/2})e^{-\alpha m^{1/2}}] + RTm^2\left(\frac{\partial C^{\phi}}{\partial P}\right)_T
$$

where

$$
{}^{\phi}V^{\text{st}} - {}^{\phi}V^{\infty} = (A_{\text{v}}/b)\ln(1 + bm^{1/2}) \qquad (2)
$$

$b = 1.2$ kg$^{1/2}$ mol$^{-1/2}$ and $\alpha = 2.0$ kg$^{1/2}$ mol$^{-1/2}$. A_{v} is the Debye–Hückel slope and the values were taken from Bradley and Pitzer (*6*). The values of the fitting parameters were obtained by nonlinear regression and are given in Tables V–VII.

Values of ${}^{\phi}V$(smoothed) at very low concentrations were calculated from the fitting parameters. The relative apparent molar volume ${}^{\phi}V^{\text{rel}}$ (${}^{\phi}V^{\text{rel}} = {}^{\phi}V$(smoothed) − ${}^{\phi}V^{\infty}$) is plotted against $m^{1/2}$ at 25 °C in Figure 1. As $m^{1/2}$ tends to zero the slope approaches A_{v}/b (*7*). Sodium and potassium hydroxides show a small positive deviation whereas lithium hydroxide has a marked negative deviation. With increasing temperature all the hydroxides show negative deviations.

The values for the apparent molar volume at infinite dilution, ${}^{\phi}V^{\infty}$, were compared where possible with existing literature data. Where sufficient data are available, the quoted results were fitted to eq 2.

(***a***) ***Lithium Hydroxide.*** A difference of 1.7 cm^3 mol^{-1} is observed between the value of −4.29 cm^3 mol^{-1} determined in

Table VI. Fitting Parameters for Sodium Hydroxide

T/K	$^{\phi}V^{\infty}$/cm^3 mol^{-1}	$10^5(\partial\beta^0/\partial P)$/ kg mol^{-1} atm^{-1}	$10^5(\partial\beta^1/\partial P)$/ kg^2 mol^{-2} atm^{-1}	$10^6(\partial C^{\phi}/\partial P)$/ kg^2 mol^{-2} atm^{-1}	std dev of fit
298.15	−4.901 ± 0.016	2.260 ± 0.118	2.991 ± 0.314	−6.792 ± 0.203	0.003
308.15	−4.154 ± 0.019	1.508 ± 0.133	3.607 ± 0.356	−5.870 ± 0.230	0.003
318.15	−3.501 ± 0.020	0.592 ± 0.139	3.982 ± 0.372	−3.718 ± 0.241	0.003
328.15	−3.389 ± 0.009	0.487 ± 0.058	3.922 ± 0.154	−3.987 ± 0.100	0.001
338.15	−3.380 ± 0.019	−0.198 ± 0.125	5.109 ± 0.334	−4.204 ± 0.216	0.003
348.15	−3.645 ± 0.011	−0.173 ± 0.070	4.549 ± 0.186	−3.248 ± 0.120	0.002

Table VII. Fitting Parameters for Potassium Hydroxide

T/K	$^{\phi}V^{\infty}$/cm^3 mol^{-1}	$10^5(\partial\beta^0/\partial P)$/ kg mol^{-1} atm^{-1}	$10^5(\partial\beta^1/\partial P)$/ kg^2 mol^{-2} atm^{-1}	$10^6(\partial C^{\phi}/\partial P)$/ kg^2 mol^{-2} atm^{-1}	std dev of fit
298.15	5.082 ± 0.020	1.592 ± 0.146	3.620 ± 0.387	−5.411 ± 0.249	0.004
308.15	5.780 ± 0.008	0.953 ± 0.055	3.767 ± 0.147	−4.054 ± 0.094	0.001
318.15	6.197 ± 0.023	0.254 ± 0.155	4.542 ± 0.413	−3.765 ± 0.265	0.004
328.15	6.254 ± 0.019	0.119 ± 0.129	4.585 ± 0.342	−4.075 ± 0.220	0.003
338.15	6.394 ± 0.023	−1.024 ± 0.146	6.350 ± 0.387	−3.304 ± 0.249	0.004
348.15	6.243 ± 0.015	−1.281 ± 0.093	5.959 ± 0.246	−1.730 ± 0.158	0.003

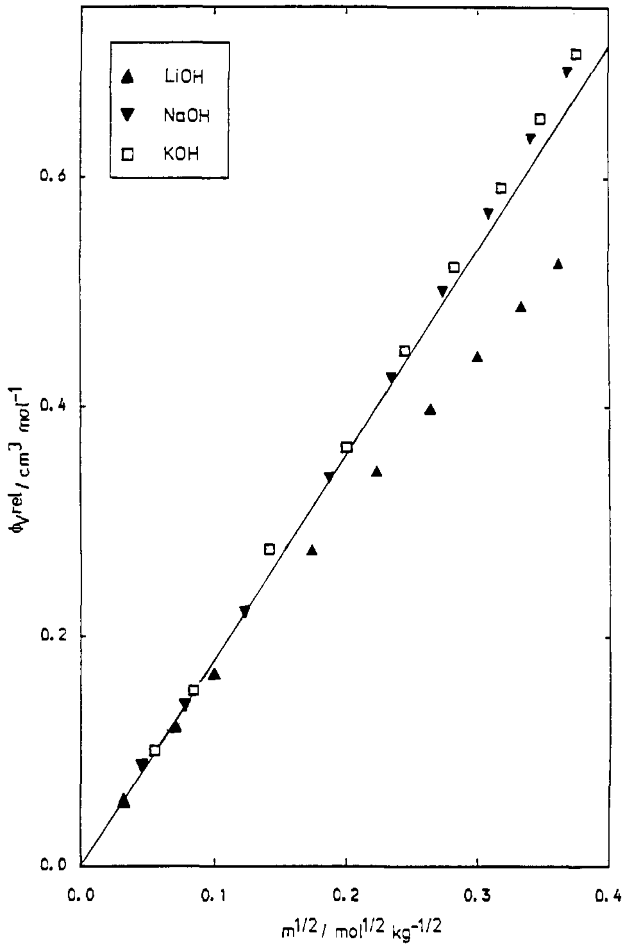

Figure 1. Comparison of $^{\phi}V^{\rm rel}$ for the alkali hydroxides at 25 °C.

Table VIII. Comparison of Smoothed Values[a] of $^{\phi}V$ for LiOH, NaOH, and KOH at 25 and 55 °C

	$^{\phi}V$/cm^3 mol^{-1}					
	25 °C			55 °C		
m/mol kg^{-1}	(a)	(b)	(c)	(a)	(b)	(c)
		LiOH				
0.0	−4.29	−5.06	−4.96	−4.33	−5.05	−5.01
0.5	−3.17	−3.02	−3.04	−3.05	−2.88	−2.87
1.0	−2.57	−2.21	−2.19	−2.44	−2.01	−2.01
1.5	−2.01	−1.61	−1.57	−1.86	−1.41	−1.42
2.0	−1.46	−1.09	−1.07	−1.30	−0.93	−0.93
2.5	−0.92	−0.61	−0.62	−0.75	−0.51	−0.50
3.0	−0.40	−0.16	−0.19	−0.21	−0.10	−0.08
		NaOH				
0.0	−4.90	−5.26	−5.46	−3.39	−3.39	−3.42
0.5	−3.28	−3.32	−3.32	−1.79	−1.62	−1.60
1.0	−2.30	−2.25	−2.20	−0.96	−0.76	−0.74
1.5	−1.44	−1.37	−1.33	−0.24	−0.04	−0.03
2.0	−0.68	−0.60	−0.58	0.41	0.61	0.60
2.5	−0.01	0.10	−0.09	0.99	1.21	1.18
3.0	0.56	0.74	0.70	1.50	1.76	1.73
		KOH				
0.0	5.08	4.92	4.83	6.25	6.25	6.17
0.5	6.58	6.74	6.79	7.79	7.99	8.01
1.0	7.48	7.72	7.78	8.58	8.80	8.83
1.5	8.29	8.52	8.55	9.29	9.46	9.49
2.0	9.03	9.22	9.21	9.92	10.05	10.07
2.5	9.68	9.85	9.80	10.49	10.60	10.59
3.0	10.27	10.42	10.34	10.99	11.10	11.08

[a] a = this work; b = ref 10 (their parameters); c = ref 10 (Pitzer parameters).

this work for $^{\phi}V^{\infty}$ at 25 °C and the value of −6.0 cm^3 mol^{-1} recorded by Gucker (*8*). Gucker's value is, however, based on the work of Lanman and Mair (*9*), who quote only three density measurements for solutions between 0.5 and 2.2 *m* and no experimental details of the method used in their determination. The lack of data made it impossible to carry out a check analysis using the Pitzer equation. Recently Roux et al. (*10*) have used the vibrating tube method to determine the density of aqueous LiOH at 25 °C over the range 0.04–3.8 *m* and at 55 °C over the range 0.1–3.8 *m*. A comparison is made of the apparent molar volumes at rounded values of the molality in Table VIII. The smoothed values (b) quoted from ref *10* were calculated from their fitting parameters for a polynomial in $m^{1/2}$. As can be seen from Table VIII, the magnitudes of the $^{\phi}V^{\infty}$ value from this work are less than theirs by 0.7 cm^3 mol^{-1} although at the higher concentrations our $^{\phi}V$ is from 0.1 to 0.3 cm^3 mol^{-1} larger at both temperatures. The data from ref 10 were also analyzed by using the Pitzer equation; these smoothed values are given in column (c) in Table VIII. It can be seen that the method of smoothing the data affects $^{\phi}V^{\infty}$ by 0.1–0.2 cm^3 mol^{-1}.

(b) Sodium Hydroxide. Reported values of $^{\phi}V^{\infty}$ for sodium hydroxide at 25 °C lie in a range from −6.80 to −4.60 cm^3 mol^{-1} and are shown in Table IX. Our value of −4.90 cm^3 mol^{-1} lies to one end of this range among the determinations made at very low concentrations. The value of Alkerlöf and Kegeles (*16*) was obtained from high-concentration data and also no measurements were actually made at 25 °C; the tabulated value was obtained by least-squares analysis from data obtained at 10-deg intervals between 0 and 100 °C. Values 7 and 8 in Table IX were both based on the work of Lanman and Mair (*9*), which consisted of only three measurements. The work of Hepler et al. (*13*) quoted enough data for analysis using the

Table IX. Values of $^{\phi}V^{\infty}$ for NaOH at 25 °C

no.	$^{\phi}V^{\infty}$/cm^3 mol^{-1}	method of measurement	concn range/ mol kg^{-1}	source
1	-4.60	dilatometer	0.006–0.65	Bodanszky and Kauzmann (*11*)
2	-4.90	weight dilatometer	0.5–3.0	this work
3	-5.19	magnetic float	0.001–1.02	Millero et al. (*12*)
4	-5.25	dilatometer	0.002–0.85	Hepler et al. (*13*)
5	-5.26	vibrating tube	0.03–10.9	Roux et al. (*10*)
6	-5.37	vibrating tube	0.19–0.37	Allred and Woolley (*14*)
7	-5.42	pressure transducer	0.01–0.15	Swarin and Curran (*15*)
8	-5.94	pycnometer	1–26	Akerlöf and Kegeles (*16*)
9	-6.48	unknown	0.5–2.2	Lanman and Mair (*9*)
10	-6.80	calcd from ref *9*		Owen and Brinkley (*17*)

Table X. Values of $^{\phi}V^{\infty}$ for KOH at 25 °C

no.	$^{\phi}V^{\infty}$/cm^3 mol^{-1}	method of measurement	concn range/ mol kg^{-1}	source
1	5.08	weight dilatometer	0.5–3.0	this work
2	4.92	vibrating tube	0.02–15.5	Roux et al. (*10*)
3	4.06	pycnometer	1–17	Akerlöf and Bender (*18*)
4	3.6	calcd from ref *18*		Fajans and Johnson (*19*)
5	3.4	no source of data quoted		Owen and Brinkley (*17*)
6	2.9	calcd from ref *9*		Gucker (*8*)

Pitzer equation: this showed perfect agreement with their reported value of -5.25 cm^3 mol^{-1}. Although our value of $^{\phi}V^{\infty}$ at 25 °C differs from that of Roux et al. (*10*) by 0.4 cm^3 mol^{-1} it can be seen from Table VIII that very good agreement is obtained for $^{\phi}V$ over the range 0.5–3.0 m; at 55 °C perfect agreement is obtained for $^{\phi}V^{\infty}$ (Table VIII) although at higher concentrations it is only ±0.2 cm^3 mol^{-1}.

(*c*) *Potassium Hydroxide*. Values of $^{\phi}V^{\infty}$ at 25 °C ranging from 2.9–4.92 cm^3 mol^{-1} have been reported and are shown in Table X. The value of 5.08 cm^3 mol^{-1} found in this work is significantly higher than the other reported values. Value No. 3 in the table is based on a least-squares analysis of data measured at 10-deg intervals between 0 and 100 °C and so no comparison of basic data is possible. Measurement 4 is reported as being based on the data of Akerlöf and Bender (*18*). No source of data is quoted for value 5. The lowest value shown, No. 6, is based on only three measurements of Lanman and Mair (*9*). Comparing $^{\phi}V$ at rounded molalities, over the experimental molality range, wtih the data of Roux et al. (*10*) (Table VIII), it can be seen that although our $^{\phi}V^{\infty}$ value at 25 °C is greater by 0.2 cm^3 mol^{-1} at higher concentrations our apparent molal volumes are less by 0.15–0.25 cm^3 mol^{-1}. Exact agreement is obtained for $^{\phi}V^{\infty}$ at 55 °C (Table VIII) but over the molality range 0.5–3.0 covered by both sets of experimental measurements, the $^{\phi}V$ values differ by 0.1–0.2 cm^3 mol^{-1}.

Glossary

A_v	Debye–Hückel slope
M_2	molar mass of solute
m	molality of solute
$^{\phi}V$	apparent molar volume
$^{\phi}V^{\infty}$	apparent molar volume at infinite dilution
$^{\phi}V^{\mathrm{rel}}$	$^{\phi}V$(smoothed) - $^{\phi}V^{\infty}$
ρ	density of solution
ρ_0	density of water

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Registry No. KOH, 1310-58-3; NaOH, 1310-73-2; LiOH, 1310-65-2.

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