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Improvement of corrosion process control techniques at engineering facilities under high parameters of water coolants [*]

V. N. Shcherbakov[1][**]

[1]Don State Technical University, Rostov-on-Don, Russian Federation

Совершенствование методов контроля процессов коррозии на объектах машиностроения при высоких параметрах водных теплоносителей[***]

В. Н. Щербаков[1][**]
[1]Донской государственный технический университет, г. Ростов-на-Дону, Российская Федерация

Introduction. The work objective is to increase the reliability of the prediction methods for the lithium hydroxide behavior in the steam-water circuit at the thermal power plants and nuclear power plants, and for the operational monitoring of the pH index of steam solutions. A method of operational control is developed on the basis of the conductometric measurements of the hydrogen index of the corrosion inhibitor vapor solutions for construction materials of lithium hydroxide used at the TPP and NPS.

Materials and Methods. A mathematical model method is used for the practical implementation of the high-temperature operational control of the steam solution pH index.

Research Results. A method for monitoring the pH of vapor solutions of lithium hydroxide based on the determination of vapor concentration through the steam condensation in the coolable conductivity sensor located in the vapor space of the steam generator is developed. This has significantly improved the accuracy of determining the lithium hydroxide concentration. Equations describing the change in the limiting molar equivalent conductance and dissociation constants of lithium hydroxide in a wide range of state change parameters are proposed.

Discussion and Conclusions. The proposed on-line technique of testing the pH value of steam solutions, and mathematical models for calculating the limiting molar equivalent conductance and dissociation constants provide an acceptable error level calculations and the capability of measurements automation. With an increase in the vapor temperature up to 573.15 K, the necessity arises to fortify lithium hydroxide in the vapor to 10^{-2} mol / kg.

Введение. Цель работы — повышение надёжности методов прогнозирования поведения гидроксида лития в пароводяном тракте на ТЭС и АЭС и оперативного контроля водородного показателя паровых растворов. Разработан метод оперативного контроля на основе кондуктометрических измерений водородного показателя паровых растворов ингибитора коррозии конструкционных материалов гидроксида лития, применяемого на ТЭС и АЭС.

Материалы и методы. Для практической реализации высокотемпературного оперативного контроля водородного показателя паровых растворов использован метод математического моделирования.

Результаты исследования. Разработан метод контроля водородного показателя паровых растворов гидроксида лития, основанный на определении концентрации в паре путём конденсации пара в охлаждаемом кондуктометрическом датчике, размещённом в паровом пространстве парогенератора, что позволило значительно повысить точность определения концентрации гидроксида лития. Предложены уравнения, описывающие изменение предельной молярной эквивалентной электропроводности и констант диссоциации гидроксида лития в широком диапазоне изменения параметров состояния.

Обсуждение и заключения. Предложенные метод оперативного контроля водородного показателя паровых растворов и математические модели для расчёта предельной эквивалентной молярной электропроводности и констант диссоциации обеспечивают приемлемую для практических расчётов погрешность и возможность автоматизации измерений. При увеличении температуры пара до 573,15 K возникает необходимость в увеличении концентрации гидроксида лития в паре до 10^{-2} моль/кг.

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[*] The research is done within the frame of independent R&D.
[**] E-mail: vladnik48@aaanet.ru
[***] Работа выполнена в рамках инициативной НИР.

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Introduction. When organizing and maintaining water chemistry regimes (WCR) at TPPs and NPPs, one of the major tasks is to minimize corrosive damages to the thermal power equipment in the zone of contact with water and steam [1]. Addition of *LiOH* in sufficient quantities into feedwater provides the creation of a hard protective lithium-ferritic film on the metal surface, which slows down corrosion and stress-corrosion cracking of the heat-exchange tubes of steam generators of NPPs with VVER-1000 [2, 3], as well as a significant increase in uptime [3]. When *LiOH* is added into the feedwater of the TG – 104 drum boilers at the TPP, the formation of a protective lithium-ferritic film is also observed [4]. *LiOH* is used at the TPPs in Russia and abroad [4–6]. In advanced fourth-generation nuclear reactors – *SCWR* (*supercritical water-cooled reactor*), providing an increase in thermal efficiency compared to the existing ones (*PWR*) from 33 to 44%, at pressure $P = 25\ MPa$, T temperature of water vapor is planned to be increased to 953 K [6]. We need data on Λ_o limiting equivalent conductance and K_d *LiOH* dissociation constants at high P and T, presented in an easy-to-calculate form, to create on-line monitoring of the *pH* values of aqueous and steam *LiOH* solutions at TPPs and NPPs that determine the effectiveness of anticorrosive protection. The technique for calculating the *pH* of *LiOH* solutions based on the data on χ electric conductivity proposed by the authors for the liquid phase [7] is unacceptable for the vapor phase at low ρ densities and C concentration due to a high error in determining χ and K_d [8].

The work objective is to increase the reliability of methods for predicting *LiOH* behavior in the steam/water circuit at TPPs and NPPs and of the operational control of the of steam solutions *pH*.

To achieve this goal, equations are developed for calculating Λ_o and K_d on the saturation line and in superheated steam at T up to 773.15 K; and a technique to control *pH* of *LiOH* vapor solutions is worked out.

Materials and Methods. Compared with the traditional control associated with sampling and cooling of samples, the operational *pH* control using conductometric sensors installed in the heat conductor of the steam generators and working at T and P close to the operating ones, is more objective and practically inertia-free [7, 8]. In *pH* calculations for the liquid phase in [7], the experimental data on K_d *LiOH* [8], K_w ionic product [9], and ε dielectric capacitance for water [10] were used. C concentration of *LiOH* aqueous solutions was determined in terms of $C\ LiOH=f\ (\chi\ LiOH)_{T=Const}$, the dependences obtained on the basis of the experimental data on χ solutions.

Under measuring χ solutions in the vapor phase (especially for small ρ and C), when the polar properties of the solvent are weakened, the dissociation degree of *LiOH* is small, and the resistance of the interelectrode space of the measure cell of the conductometric converter can become commensurate with the resistance of the electrical insulator, which will lead to large errors in determining χ [8].

For the practical implementation of the high-temperature operative *pH* control, the necessary data on Λ_o and K_d can be obtained in the studied range of the state parameter variation through the mathematical simulation technique and be presented in the form of equations.

Research Results. Considering the analysis of existing methods, this paper proposes a method for testing *pH* of *LiOH* vapor solutions based on the determination of *C LiOH* in steam through the vapor condensation in the conductivity sensor with a cooled capillary [11] placed in the vapor space of the steam generator in the coolant flow. In this case, *C LiOH* in steam is equal to *C LiOH* in the vapor condensate and can be determined from $C\ LiOH=f\ (\chi\ LiOH)_{T=Const}$ dependences obtained for the liquid phase [7]. Vapor condensation can significantly improve the accuracy of determining χ and C [8]. The *pH* calculations for *LiOH* vapor solutions were performed according to the procedure described in [12] using data from [8] on K_d.

To assess the effectiveness of using *LiOH* for corrosion protection in *SCWR* fourth-generation atomic reactors, the authors [6] calculated the *pH* of *LiOH* aqueous and steam solutions at $P = 25\ MPa$ in a wide variation range of ρ, T, C, on the basis of the experimental data on K_d [13–15]. Besides, an empirical equation for calculating K_a molal Kon constant [16] in ρ variation range from 50 to 1000 kg / m^3 and T from 373 to 873 K is proposed:

$$log_{10}K_a = a_1 + a_2/\tau + + (a_3 + a_4/\tau)\cdot log_{10}\delta + (a_5 + a_6/\tau)\cdot(log\delta)^2,$$

where $\delta=\rho/\rho_c$; $\tau=T/T_c$; ρ_c, T_c is the solution density and temperature at the critical point; a_1, a_2, a_3, a_4, a_5, a_6 are constants.

In superheated steam, with a decrease in ρ and *T* K_d *LiOH* reduces, the difference between *pH H2O* and *pH* of *LiOH* steam solutions decreases, which reduces the effectiveness of corrosion protection of the structural elements at high *P* and *T*. The authors have calculated the *pH* values of *LiOH* steam solutions at *P = 25 MPa*; *T* = 673, 723 and 773 *K* for *C*=10^{-6}, 10^{-5}, 10^{-4}, 10^{-3} and 10^{-2} mol/kg. The experimental data on K_d *LiOH* [8], K_w ionic product [9], and ε dielectric capacitance of water [10] were used in the calculations. Fig. 1 shows *pH* dependence of aqueous and vapor *LiOH* solutions on ρ at *P = 25 MPa* for *C*=10^{-6}–10^{2} mol/kg.

In the area of ρ <330 kg/m^3, the authors [13–15] did not perform experimental studies; therefore, the figure shows only the results of calculating *pH* of *LiOH* using the data on K_d *LiOH* obtained by the authors' equation [16].

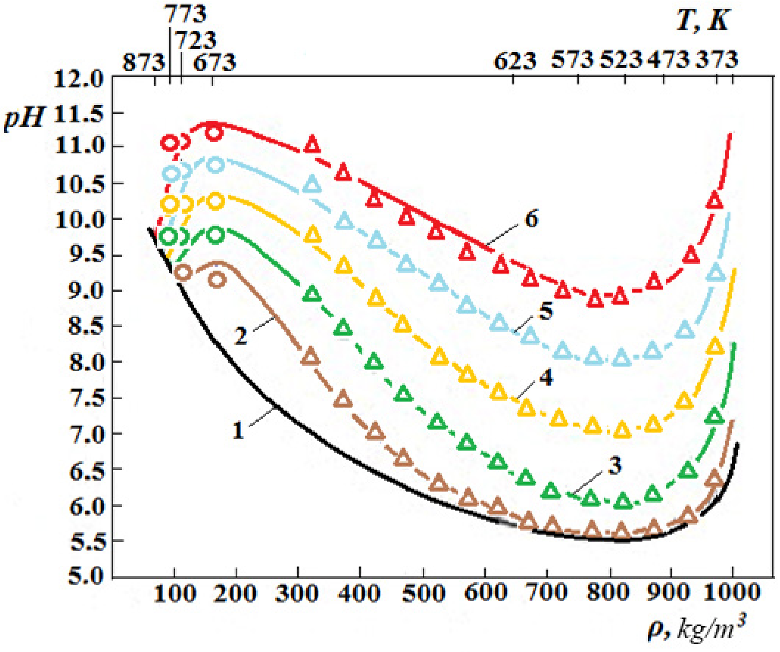

Fig.1. *pH* dependence of *LiOH* aqueous and steam solutions on ρ density at *P* = 25 *MPa*: 1 – H_2O; 2,3,4,5,6 – *C LiOH* = 10^{-6}, 10^{-5}, 10^{-4}, 10^{-3}, 10^{-2} mol/kg; Δ, ○ is calculation of *pH* from data on K_d [13-15] and [8]; 2,3,4,5,6 lines are calculation of *pH* according to K_d data obtained from equation [16].

Λ_o and K_d values [8], which are necessary for creating equations describing the change of Λ_o and K_d with changing ρ and *T*, are given in Tables 1 and 2 in comparison with the data from other authors. These values were previously obtained by the authors on the basis of the measurements of the electrical conductivity of high-temperature *LiOH* solutions.

To create equations describing Λ_o behavior on the saturation line, in order to reduce the error of data approximation, ρ variation range was divided into 2 parts (from 100 to 500 and from 500 to 958 kg/m^3), for each of which an equation of the following form was proposed:

$$\Lambda_o=b_o\cdot\rho^3 + b_1\cdot\rho^2 + b_2\cdot\rho + b_3,$$

where b_o, b_1, b_2, b_3 are definable constants; ρ is density of the solution, kg/m^3. b_o, b_1, b_2, b_3 values for ρ = (100–500) and (500–958) kg/m^3 are –0.000004; 0.005878;

–2,810,820; 1942.943226 and –1.25711 10^{-5}; 0.024177411; –16.58559801; 5334,222549 respectively. For pK_d=–log_{10} values, the values of d_1–d_6 coefficients are calculated in the equation similar to that presented in [16] and given above:

$$pK_d=d_1 + d_2/\tau + (d_3 + d_4/\tau)\cdot log_{10}\delta + (d_5 + d_6/\tau)\cdot(log\delta)^2,$$

where $\delta=\rho/\rho_c$; $\tau=T/T_c$; ρ_c, T_c are density and temperature of the solution at the critical point; d_1, d_2, d_3, d_4, d_5, d_6 are constants.

When calculating d_1–d_6 constants, we used previously obtained experimental data on K_d [8]. On the saturation line, we considered only our own experimental data. Calculations for the superheated steam at *T* = 673.15 and 773.15 *K* were made in two versions: in the first one, only the author's data on K_d [8] were considered, in the second – all the data given in Table 2.

Table 1

Dependence of Λ_o *LiOH* limiting molal conductivity on ρ density
on saturation line and in superheated steam

ρ, kg/m^3	$\Lambda_o \cdot 10$, cm·m^2 / kmol		
	Saturation line	T = 673 K	T = 773 K
100	1710[8]	1675[8]	1665[8]
130	1675[8]	1630[8]	1620[8]
160	1630[8]	1590[8]	1575[8]
200	1575[8]	1550[8]	1545[8]
300	1510[8]	1480[8]	1475[8]
400	1475[8]	1460[8]; 1392[13]	1455[8]
433		1452[15]	
500	1450[8]	1435[8]; 1539[13]	1428[8]; 1574[13]
600	1370[8]	1380[8]; 1355[13]	1388[8]; 1377[13]
621	1277[15] (P = 23.86 MPa)	-	-
700	1265[8]	1275[8]; 1270[13]	1276[13]
714	1189[15] (P = 9.52 MPa)	-	-
800	1100[8]	1088[13]	1109[13]
871	1017[15] (P = 9.91 MPa)	-	-
917	780[8]	-	-
922	754[15] (P = 9.4 MPa)	-	-
950	594[13]	-	-
958	573[8]	-	-

For the saturation line: $d_1=119.50984$; $d_2=-115.33347$; $d_3=-174.20233$; $d_4=167.90533$; $d_5=-143.78368$; $d_6=183.09356$.

For the first version of the calculation for the superheated steam: $d_1=7.6323975$; $d_2=-3.214114$; $d_3=-10.9289$; $d_4=2.5463453$; $d_5=-10.34132$; $d_6=15.190972$; for the second version of the calculation: $d_1=10.113404$; $d_2=-6.133638$; $d_3=-17.85984$; $d_4=8.888042$; $d_5=-8.405401$; $d_6=14.648395$.

The percent deviations of the experimental values of Λ_o and pK_d from those calculated by the proposed equations are presented in Table 3.

Table 2

Dependence of negative logarithm of Koff constant of *LiOH* pK_d
on ρ density on saturation line and in superheated steam

ρ, kg/m^3	pK_d, mol/kg		
	Saturation line	T = 673.15 K	T = 773.15 K
100	10.04[8]	10.07[8]; 11.06 [16]	10.16[8]; 13.34[16]
130	8.41[8]	8.56[8]	8.67[8]
160	7.24[8]	7.32[8]	7.69[8]
200	6.25[8]	6.42[8]; 6.45[16]	6.89[8]; 8.16[16]
300	4.40[8]	4.69[8]; 4.13[16]	5.20[8]; 5.64[16]
379		3.48[15]	
400	3.86[8]	3.98[8]; 2.82[16]; 3.14[13]	4.31[8]; 4.01[16]
433		3.1[15]	
500	3.28[8]	3.38[8]; 2.81[13]	3.43[8]; 3.15[13]
600	2.52[8];	2.45[8]; 2.32[13]; 1.26[16]	2.58[8]; 2.43[13]; 1.97[16]
650		2.13[13]	2.25[13]
700	2.08[8];	2.01[8]; 1.83[13]	2.07[13]
712	1.59[17]		
750		1.82[13]	1.89[13]
800		1.63[13]; 0.343[16]	1.65[13]; 0.753[16]
807	1.41[15] (P = 11.02 MPa)	-	-
922	1.29[15] (P = 9.4 MPa)	-	-

961	1.03[15] ($P = 4.88$ *MPa*)	-	-

Table 3

Deviation (in%) of experimental values of limiting equivalent conductance ($\delta\Lambda_o$) and negative logarithm of molal Koff constants (δpK_d) from values calculated by equations

ρ, kg/m^3	$\delta\Lambda_o$	δpK_d				
	Saturation line	Saturation line	T=673,15 K 1 calculation version	T=773,15 K 1 calculation version	T=673,15 K 2 calculation version	T=773,15 K 2 calculation version
100	0.36[8]	0.04[8]	1.20[8]	1.36[8]	3.40[8]; 5.86 [16]	10.9[8]; 15.5[16]
130	0.47[8]	0.23[8]	0.18[8]	1.22[8]	2.42[8]	11.7[8]
160	0.28[8]	0.33[8]	2.62[8]	1.86[8]	3.42 [8]	10.5[8]
200	0.34[8]	0.11[8]	0.92[8]	0.39[8]	0.52[8]; 0.98[16]	5.95[8]; 10.5[16]
300	0.09[8]		2.50[8]	0.29[8]	3.86[8]; 9.17[16]	1.99[8]; 5.96[16]
379					3.05[15]	
400	0.03[8]	0.12[8]	4.99[8]	4.07[8]	14.8[8]; 20.2[16]; 7.96[13]	6.44[8]; 0.56[16];
433					0.41[15]	
500	0.09[8]	0.16[8]	8.95[8]	2.40[8]	21.77[8]; 5.90[13]	8.78[8]; 0.67[13]
600	0.10[8]	0.17[8]	4.56[8]	6.11[8]	13.72[8]; 8.89[13]; 67.8[16]	5.24[8]; 0.60[13]; 24.1[16]
650	-	-	-	-	10.6[13]	4.02[13]
700	0.44[8]	0.05[8]	7.82[8]	-	14.38[8]; 5.95[13]	8.03[13]
750		-	-	-	14.2[13]	11.5[13]
800	0.26[8]	-	-	-	12.8[13]	11.3[13]
917	2.27[8]	-	-	-	-	-
958	1.51[8]	-	-	-	-	-

Discussion and Conclusions. As obvious from Fig. 1, pH values of $LiOH$ obtained by the authors at $T = 673$ K are lower compared to those obtained through calculating [6] by a mean of 0.2; at $T = 723$ K, they are higher by 0.1–0.2; at $T = 773$ K, they are higher by 0.1–0.7 pH units.

Thus, the error ratio of the data on pH of $LiOH$ in the superheated steam obtained by the authors and presented in [6] goes up with increase in T and C. Such a mismatch may be considered satisfactory if it is remembered that with decreasing ρ to 200–100 kg/m^3, the experimental data [8] error on K_d $LiOH$ increases to 180%, and, moreover, the authors [6] did not use the experimental data on K_d equation at $\rho < 330$ kg/m^3.

The deviation of the experimental values of Λ_o and pK_d from those calculated using the equations proposed in this paper on the saturation line does not exceed 1%, and only at $\rho = 917$ and $\rho = 958$ kg/m^3, it reaches 2.27 and 1.51% for Λ_o (Table 3). In the superheated steam at $T = 673.15$ and $T = 773.15$ K for the first version of the calculation using only the author's experimental data on K_d [8], the greatest deviation of the experimental data of pK_d from the calculated one does not exceed 2.5% at $\rho = 100$–300 kg/m^3 and goes up to 8.95% with an increase in ρ to 400–700 kg/m^3. For the second version of the calculation, considering the data of all authors presented in Table 2, the greatest error of calculation by the equation at $\rho = 100$–300 kg/m^{3}and $T = 673.15$ K; $T = 773.15$ K reaches 15.5% for these authors [16], and at $\rho = 100$–300 kg/m^3 and $T = 673.15$ K; $T = 773.15$ K – 67.8%.

Based on the analysis of the obtained results, it can be concluded that the use of $LiOH$ as a corrective additive for pH control in order to prevent corrosion damage to structural elements of *SCVR* advanced atomic reactors of the fourth generation creates no difficulties at $T \leq 673.15$ K and $P = 25$ MPa. With an increase in temperature already at $T = 773.15$ K, for effective corrosion protection, it is necessary to raise C $LiOH$ to 10^{-2} mol/kg, which puts forward the task to study the vapor solvability of $LiOH$ at these state parameters.

The proposed method of operative control of *pH* of $LiOH$ vapor solutions and mathematical models for calculating Λ_o and K_d provide the capability to determine *pH* using a measuring-computing complex that runs continuously in automatic mode; it has a short signal delay time, and it provides an acceptable for practical calculations error of measurement results.

The application of such devices in modern systems of chemical-engineering monitoring at TPPs and NPPs will significantly improve the reliability of *pH* operation control and forecasting techniques of $LiOH$ behavior of in the vapor-water circuit.

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Author:

Shcherbakov, Vladimir N.,
associate professor of the Hydraulics, Hydraulic and
Pneumatic Control Systems and Heat Processes
Department, Don State Technical University
(1, Gagarin sq., Rostov-on-Don, 344000, RF),
Cand.Sci. (Eng.), associate professor,
ORCID: http://orcid.org/10000-0001-5682-3792
vladnik48@aaanet.ru