



Linear transformations of the Butler–Volmer equation

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ABSTRACT

The conventional ways of determining the kinetic parameters (exchange or corrosion current and Tafel slopes) of the Butler–Volmer (BV) equation have serious drawbacks. In this paper linear transformations of the BV equation are proposed, based on the observation that the BV equation is a solution of a second-order, linear, homogeneous differential equation. The proposed method offers a very simple multilinear calculation method which can be applied in narrower or wider polarization ranges around the equilibrium or corrosion potential. The performance of a linearized form was tested on simulated and measured data series.

1. Introduction

Assuming that the charge transfer is the rate-determining step, the current vs. potential relationship of an electrochemical reaction can be given via the following equation:

$$J = J_0 \left(\exp \left[\frac{E_p - E_0}{b_A} \right] - \exp \left[-\frac{E_p - E_0}{b_C} \right] \right), \quad (1)$$

where J is the current density, E_p is the polarized potential and b_A is the anodic and b_C is the cathodic Tafel slope. The corresponding decadic Tafel slopes are $b_{A,10} = b_A \cdot \ln 10$ and $b_{C,10} = b_C \cdot \ln 10$, respectively. In simple redox systems E_0 is the equilibrium potential, J_0 is the exchange current density and $E_p - E_0$ is the overvoltage. For redox systems Equation (1) is called the Butler–Volmer (BV) equation [1]. In corrosion systems (mixed kinetic systems with more than one redox reaction) E_0 is the corrosion potential, J_0 is the corrosion current density and the value $E_p - E_0$ is called polarization. As the experimental systems discussed below are typical corrosion systems, we shall use the corrosion terminology, but the whole discussion is valid for all systems which obey the BV equation. Equation (1) can be written in more complex forms [2,3,4], if the pre-exponential factor is not constant, typically due to transport effects, but in this paper we do not deal with this case. For corrosion systems Equation (1) is sometimes called the Wagner–Traud equation [5], but it is also called the BV equation ([6], section 4.2.2). For clarity and brevity, we shall refer to Equation (1) as the BV equation. For the same reasons the substitution $\eta = E_p - E_0$ will be introduced, both for the notation of the overvoltage and the polarization.

The practical application of the BV equation is severely complicated by the difficulties of determining the parameters J_0 , b_A and b_C , because nonlinear parameter fitting, which can be applied in principle in the whole polarization range, has a number of drawbacks and problems. Its application needs rather accurate initial values and, even if they are provided, the assessed parameters are ambiguous; one can never be sure that really the best approximation of the parameters is found. Also, nonlinear parameter fitting algorithms need more sophisticated and expensive statistical software packages. Due to these disadvantages, nonlinear parameter fitting is avoided whenever possible. Not surprisingly, the most popular method of assessing the parameters is the linear fitting of the $\lg J$ vs. η curve in the ranges $\eta > b_A$ or $\eta < -b_C$. However, in this method only the data range of greater overvoltage/polarization is used, and in this range the parameters may be different to those effective in the vicinity of the equilibrium/corrosion potential because the current vs. potential relationship can be corrupted by transport limitations or change of mechanism. Alternatively, in corrosion systems, the corrosion rate can be assessed via the value of the polarization resistance, obtained from electrochemical impedance spectroscopy (EIS) or DC measurements. However, the values of the Tafel slopes are necessary for the calculation of the corrosion current from the value of the polarization resistance. A somewhat oversimplified, but frequently applied solution of the problem, especially in industrial corrosion monitoring systems (linear polarization resistance, LPR measurement) is that, instead of the experimental determination, the Tafel slopes are substituted with hypothetical values on an empirical basis. Reviewing experimental surveys on DC polarization measurements suggest that the Tafel slopes can vary in a wide range from about 40 mV/decade up to a few hundred mV/decade, even under very similar conditions

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[7–14], which underlines the necessity of the assessment of the Tafel slopes in order to achieve an acceptably accurate corrosion rate determination.

In what follows, we present a new method of linear transformation of the BV equation, applicable over the whole potential range, which can substantially simplify the task of accurate parameter determination.

2. Theoretical discussion

From the standard textbooks of higher mathematics and mathematical physics (e.g. [15,16]) it is well known that the general solution of a second-order, linear, homogeneous differential equation with constant coefficients can be written as follows:

$$y(x) = C_1 \exp[\lambda_1 x] + C_2 \exp[\lambda_2 x]. \quad (2)$$

With appropriately chosen constants (C_1 and C_2) and parameters (λ_1, λ_2) this is identical to the BV equation. This observation suggested the idea of trying to find the differential equation which corresponds to the solution, specifically meeting the requirements of the equivalence of parameters. The resulting differential equation is the following (the detailed steps leading from Equation (1) to Equation (3) are included in the [Supplementary Information](#) (SI); note that derivatives with respect to η are, in fact, derivatives with respect to E_p (see Eq. (1)), as the two quantities differ only in the constant E_0):

$$\frac{d^2 J}{d\eta^2} = \left(\frac{1}{b_A} - \frac{1}{b_C} \right) \frac{dJ}{d\eta} + \frac{J}{b_A b_C}. \quad (3)$$

That is, the BV equation is transformed into a second-order, linear, homogeneous, complete differential equation. The BV equation is, not surprisingly, one of the solutions of this differential equation, therefore Equation (3) will be referred to henceforth as the differential form of the BV equation. To the best of our knowledge, this feature of the BV equation has not yet been mentioned in the literature. Equation (3) has two parameters, the asymmetry factor $1/b_A - 1/b_C$ and the symmetry factor $1/(b_A b_C)$, and it does not include explicitly the value of the parameter J_0 . This suggests that the equation is invariant to the exchange/corrosion current density. This is formally true, because all three variables (the current density and its derivatives) are multiplied by J_0 . However, J_0 can be calculated accurately using the values of the Tafel slopes and the value of the polarisation resistance R_p , obtained from a polynomial fitting of the current vs. potential curve in the range close to the origin. The asymmetry parameter was defined in a recent publication [17], but the symmetry parameter was defined there in a different way. This difference is attributed to the different mathematical approaches used; in the cited work the symmetric parameter was derived from a Taylor expansion of the hyperbolic sine function and in the present work it is obtained from two differentiations and several other transformations of the BV equation.

Equation (3) is a bilinear one, which can be fitted quite easily using most statistical analysis packages. However, it has a severe problem from the point of view of practical application: it requires the first and also the second derivative of the current with respect to the potential. As the derivation magnifies the effect of the measurement noise in the deviation of the data from the theory, the application of the derivatives inherently decreases the accuracy of the assessed parameters. It is possible to eliminate the differentials from Equation (3) in two integration steps. The integration of Equation (3) gives.

$$\frac{dJ}{d\eta} = \frac{1}{b_A b_C} \int_0^\eta J d\epsilon + \left(\frac{1}{b_A} - \frac{1}{b_C} \right) J(\eta) + \frac{1}{R_p}, \quad (4)$$

because the last term in Equation (4) is the integration constant; for $\eta = 0$ the left side of the equation is equal to $dJ/d\eta|_{\eta=0} = 1/R_p$ and the

first two terms on the right side are equal to zero. Since $1/R_p = J_0(1/b_A + 1/b_C)$, the J_0 dependence appears again here through the integration constant. Another integration yields:

$$J(\eta) = \frac{1}{b_A b_C} \int_0^\eta \int_0^\epsilon J d\epsilon d\epsilon + \left(\frac{1}{b_A} - \frac{1}{b_C} \right) \int_0^\eta J d\epsilon + \frac{\eta}{R_p}. \quad (5)$$

In this step the integration constant is zero because $J = 0$ if $\eta = 0$. Note that the integration variables and/or integration limits ϵ and ϵ are of the same dimension and carry the same physical content as η . From the above two forms the polarization resistance is directly obtainable. Equation (5) will be named henceforth the linearized BV equation or the integral form of the BV equation. In contrast to the differential form, the integral form absorbs and balances the random errors and is therefore much more suitable in practical calculations.

Considering that the value of the polarization resistance can be obtained separately from the linear or polynomial fitting of the points near to the equilibrium or corrosion potential, the last term of Equation (5) contains only known elements and therefore it can be rearranged to obtain a simple linear relationship of the form of $y = \text{constant}1 \cdot x + \text{constant}2$:

$$\frac{J(\eta) - \eta/R_p}{\int_0^\eta J d\epsilon} = \frac{1}{b_A b_C} \frac{\int_0^\eta \int_0^\epsilon J d\epsilon d\epsilon}{\int_0^\eta J d\epsilon} + \left(\frac{1}{b_A} - \frac{1}{b_C} \right). \quad (6)$$

The disadvantage of this equation is that the points around $\eta = 0$, where the integral in the denominator is also zero, cannot be used for evaluation purposes. However, the technical simplicity of the parameter assessment can compensate for the absence of this range of points in some cases. In this sense, Equation (6) is a simple alternative to the conventional semilogarithmic ($\lg J$ vs. η) linear fitting and extrapolation, but with the advantage of utilizing the information from the η range typically between 20 and 60 mV. Furthermore, in the ideally symmetric case, i.e., if $b_A = b_C = b$, the division by the first integral is unnecessary and a very simple function of the form of $y = x/b^2$ is obtained. During testing of these transforms, we obtained better fitting results with Equation (5), and therefore we used this in the present paper. Applicability studies of other linear transformations of Equations (3) and (5) are ongoing.

It has to be stressed that the integration of a differential equation provides the solution of it only in some special cases and this is not that case. This becomes obvious if one tries to express explicitly $J(\eta)$ from any of Equations (4) – (6). Equations (3)–(6) are pure mathematical transformations of Equation (1), which means that these equations fit the current vs. potential correctly if and only if the kinetics of the system obeys Equation (1). However, this correspondence does not give any information about the physical nature of the equations, which is different for redox and corrosion systems. In conventional discussions of corrosion kinetics, the contribution of the cathodic branch of the metal/metal ion reaction and the anodic process of the balancing oxidizing reaction(s) are generally considered negligible, although they are not necessarily that small. Making use of the linear character of the proposed equations, there is the opportunity to build more complex models which also take these processes into account.

In the following sections, the performance of Equation (5) will be demonstrated on simulated and experimental polarization curves.

3. Experimental

Synthetic data with $(2 + 2)$ % superimposed random errors (2 % evenly distributed absolute errors of J_0 plus 2 % normally distributed relative errors of J), unless otherwise stated, were generated using the parameters $J_0 = 1$ mA, $b_{A,10} = 60$ mV and $b_{C,10} = 120$ mV (general case) and also $b_{A,10} = b_{C,10} = 120$ mV (symmetric case). The extent of error applied here is larger than that of typical potentiostatic experiments. The generated points were taken equidistantly every 1 mV in the full applied η range.

In order to facilitate a direct comparison of the linearized BV equation (model LBV) with other models, experimental data from a recent paper [17] reporting a quadratic approximation model (model QA) were applied for parameter assessment.

The experimental conditions are included in Table 1. All measurements were carried out with a METROHM AUTOLAB PGSTAT 302 N type potentiostat using NOVA 1.11 software, starting at the corrosion potential with a sweep first in the negative and then in the positive direction.

The evaluation procedure was carried out in Excel, starting with the calculation of the ohmically compensated potentials with the formula $\eta = E_m - JR_\Omega - E_0$, where E_m is the measured potential data and R_Ω is the specific (Ωcm^2) ohmic solution resistance, which was calculated from EIS data. The integrals of the current were calculated with the trapezoid method. The parameters of the original BV equation ($J_0, 1/b_A, 1/b_C$, see Equation (1)) were re-calculated from the parameters of the multilinear fitting ($1/b_A - 1/b_C, 1/b_A b_C, 1/R_p$, see Equation (5)). The fitted current density data were also calculated via Equation (5). The fitting was carried out in two polarization ranges; over a wide range (i.e., the whole range of the polarization) the parameters obtained with model LBV were compared to the parameters obtained via the conventional semilogarithmic linear fitting and extrapolation. The results of the fitting in a narrow range (-25 mV ... +25 mV) were compared to the same parameters obtained in the referenced publication [17] in the same polarization range.

4. Results and discussion

The results of the fitting of the simulated data are shown in Fig. 1 and the obtained parameters are included in Table 2. The fitting of the data series shows that the model of Equation (5) is very robust even if extremely high random errors are superimposed on the model data. The input BV parameters can be obtained relatively accurately, with very good correlation coefficients of the multilinear fitting (r^2).

The polarization data for both HCl and NaCl experimental systems were fitted in two ranges. In the wide range the parameters obtained via Equation (5) were compared with those obtained from the conventional semilogarithmic, linearized fitting. This latter method requires a polarization range $\eta \gg b_A, b_C$, where there is a chance to get linear sections in this semilogarithmic representation. (Alternatively, if a nonlinear parameter fitting is applied, then it requires approximately the same range of polarization because otherwise the correlations between

Table 1

Electrolyte composition and electrode material of the experimental systems.

System code	Electrolyte	Electrode material	Sweep rate/(mV/s)	Maximum positive polarization/mV ⁽¹⁾	Maximum negative polarization /mV ⁽¹⁾
HCl [17]	1% HCl	DIN ST-52 steel	1	80	-183
NaCl [17]	5% NaCl + 0.5% acetic acid	DIN ST-52 steel	1	86	-191

⁽¹⁾ Limits obtained after the ohmic drop compensation.

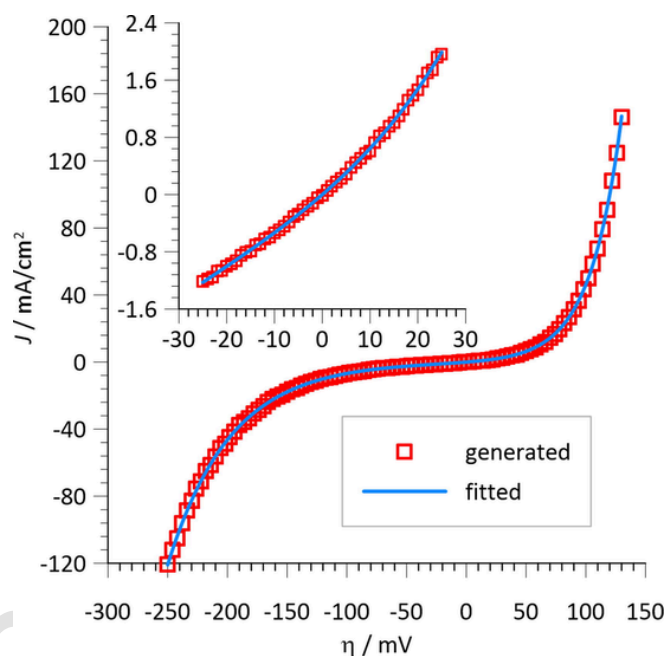


Fig. 1. Synthetic and fitted polarization curves according to Table 2. The wide polarization range curve corresponds to the fourth data column in Table 2 while the inset corresponds to the first data column in Table 2.

Table 2

Fitted parameters of the synthetic polarization curves according to Equation (5) ($b_{A,10} = 60$ mV, $b_{C,10} = 120$ mV and $J_0 = 1$ mA; for the symmetric case, $b_{A,10} = b_{C,10} = 120$ mV). $(2 + 2)$ % superimposed random errors were applied, except for the case indicated.

Parameter	Polarization range				
	± 25 mV	± 25 mV ⁽¹⁾	± 60 mV	from -250 mV to +130 mV	from -250 mV to +250 mV ⁽²⁾
$R_p / \Omega\text{cm}^2$	17.25	17.35	17.36	17.42	25.93
$b_{A,10} / \text{mV}$	62.3	60.0	59.9	60.0	120.4
$b_{C,10} / \text{mV}$	130.9	120.6	120.3	120.1	120.2
$J_0 / \text{mA/cm}^2$	1.062	1.002	1.001	0.997	1.007
r^2	0.99970	0.99997	0.99990	0.99991	0.99988

⁽¹⁾ $(1 + 0)$ % superimposed random errors.

⁽²⁾ symmetric case.

the parameters are very strong which causes degeneration of the Hesse matrix and also causes inaccuracy in other methods, too.) On the other hand, it is obvious that if the value of the corrosion current (or any other parameter) is to be assessed **at the corrosion potential**, then the optimal choice is a method that can assess the parameters from measurement data bound to a polarization range as narrow around the corrosion potential as possible, because this range will be the least biased by any additional effect related to the polarization (e.g., transport processes). Therefore, it is especially important that the model LBV can be used, theoretically, over the full polarization range (with the exception of a narrow range, say, only a few millivolts around the corrosion potential, where practically it will degenerate without the support of the points of the adjacent ranges and becomes insensitive to the individual parameters). In fact, the model LBV is applicable in the range where the curvature of the current vs. potential relationship differs sufficiently from linearity, which is generally achieved with a polarization of a few tens of millivolts. Luckily, the method of quadratic approximation (QA), published recently [17], was developed to operate in exactly this polarization range, thus providing a straightforward option for comparison.

The parameters of the fitting with the different methods are included in Table 3 and in Fig. 2 for the experimental systems. The results are significantly different for the two systems. For the HCl system, the fitting to Equation (5) is almost perfect with correlation coefficients better than “three-nine” in both the wide and in the narrow polarization ranges (see the two lower charts in Fig. 2). This result is in agreement with the observation that in the model BV (see the most upper chart in Fig. 2 and the lower rows in Table 3) the anodic and the cathodic slope extrapolations give the same value for the corrosion current and, consequently, the system can be considered to be in compliance with the BV equation. Also, the Tafel slopes obtained from the models BV (Equation (1)) and LBV (Equation (5)) fall very close to each other. The values of J_0 are somewhat different, but this can be attributed to the basically different ways of calculation. From these observations, it can be concluded that in the wide polarization range the HCl system adequately obeys the BV equation. However, the data for the narrow polarization range give different results. The parameters obtained with models QA and LBV are closely matching, but the J_0 values obtained with both methods show a larger difference than the values obtained in the wider polarization range. This is an unexpected result, indicating that corrosion kinetic parameters obtained from wide and narrow polarization ranges can show significant differences. With the introduction of the QA and LBV methods, two powerful new tools are provided for investigating this phenomenon.

For the NaCl series the conventional fitting provided different corrosion current density data from the anodic and from the cathodic polarization curves (see Table 3 and the upper chart in Fig. 2), which is the

Table 3

Kinetic constants obtained for the experimental systems with the LBV, conventional BV and quadratic approximation methods.

System code	Method	Parameter	Maximum polarization / mV		
			-200 ... +100 ⁽¹⁾	±25 ⁽²⁾	
HCl	Equation (5)	$b_{A,10}/\text{mV}$	62.5	76.5	
		$b_{C,10}/\text{mV}$	83.9	92.2	
		$J_0/$ mA/cm^2	0.0231	0.0373	
		$R_p/$ Ωcm^2	675	487	
		r^2	0.99949	0.99999	
		Semilogarithmic BV parameter fitting (Equation (1))	$b_{A,10}/\text{mV}$	63.4	N/A
		$b_{C,10}/\text{mV}$	85.3		
		$J_0/$ mA/cm^2	0.027 ⁽³⁾	0.027 ⁽⁴⁾	
	Quadratic approximation (QA) [17]		$b_{A,10}/\text{mV}$	N/A	76.7
			$b_{C,10}/\text{mV}$		92.1
			$J_0/$ mA/cm^2		0.0373
	NaCl	Equation (5)	$b_{A,10}/\text{mV}$	42.2	65.8
$b_{C,10}/\text{mV}$			145.4	99.6	
$J_0/$ mA/cm^2			0.0393	0.0285	
$R_p/$ Ωcm^2			361	604.6	
r^2			0.97820	0.99995	
Semilogarithmic BV parameter fitting (Equation (1))			$b_{A,10}/\text{mV}$	45.2	N/A
		$b_{C,10}/\text{mV}$	159.9		
		$J_0/$ mA/cm^2	0.015 ⁽³⁾	0.049 ⁽⁴⁾	
Quadratic approximation (QA) [17]			$b_{A,10}/\text{mV}$	N/A	58.1
			$b_{C,10}/\text{mV}$		87.4
			$J_0/$ mA/cm^2		0.031

(1) Polarization limits applied during the measurement; evaluation is based on the ohmically compensated potential values which comprise narrower ranges.

(2) Ohmically compensated polarization range.

(3) Calculated from the anodic polarization curve.

(4) Calculated from the cathodic polarization curve.

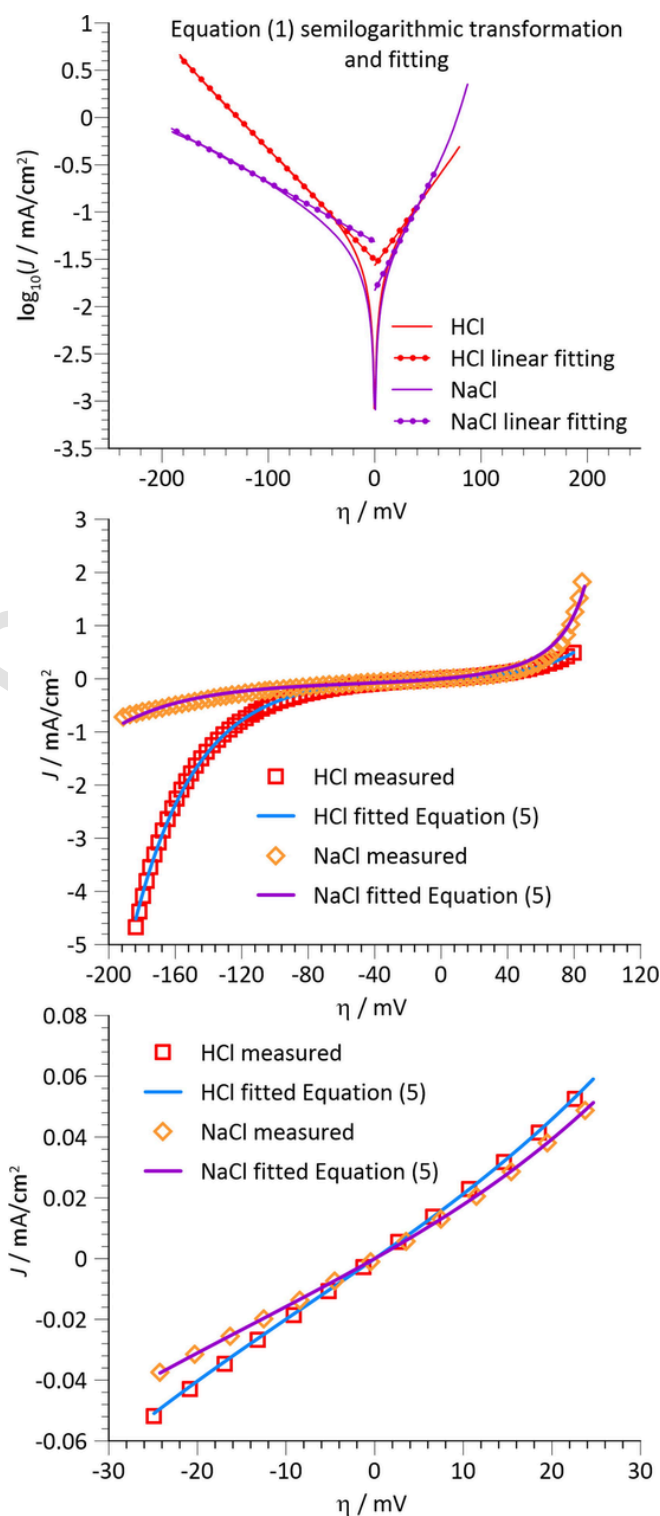


Fig. 2. Experimental measurement curves and their fitting with the semilogarithmic BV transformation (above) and with Equation (5) in the wide and in the narrow polarization ranges.

sign of an anomaly (from this methodological point of view, it is irrelevant whether the anomaly comes from a change of mechanism or from some problem in the measurement). Not surprisingly, the corresponding fitting with Equation (5) is also much worse for the wide polarization range than that of the HCl series. However, in the narrow polarization range the QA and the LBV methods give similar kinetic parameters and in this narrow range the correlation coefficient of the fitting is also

extremely good. This result underlines the necessity and usefulness of the application of evaluation methods for corrosion kinetic parameters using only a narrow polarization range around the corrosion potential.

5. Conclusions

In an effort to provide a linearized form of the BV equation, it was observed that the BV equation was, in fact, the solution of a constant coefficient, linear, homogeneous, complete second-order differential equation. With the appropriate transformations, the theorem was proven and the coefficients were determined. As the obtained differential equation contains the first and second derivatives of the current with respect to the potential, and the calculation of them typically involves a significant level of noise in the measurement data, the differential form of the LBV equation was integrated twice and the resulting linear integral equation was used for the assessment of the electrochemical kinetic parameters (Tafel slopes and corrosion/exchange current density). The method was tested on error-superimposed synthetic data series which returned the input kinetic parameters with high accuracy, proving the robustness of the applied method. Furthermore, two experimental systems, previously used for the testing of another new parameter assessment method (model QA) were applied for the testing and evaluation of the performance of the model LBV. For the HCl system, which apparently correctly obeyed the BV kinetic equation, the comparison of the kinetic parameters obtained with the different methods confirmed that the LBV model returned approximately the same kinetic parameters as the conventional BV model. However, the corrosion current obtained by fitting to a narrower polarization range was significantly different. This observation highlights the fact that excessive polarization may change the mechanism or, at least, the values of the kinetic parameters, which must be taken into account in electrochemical corrosion experiments and, particularly, industrial corrosion monitoring. In summary, the proposed linear transformations of the BV equation provide significant advantages over conventional methods for the assessment of the BV parameters. These interesting results were born from a new way of looking at a basic electrochemical equation, and we believe that it is worth considering their future applications.

CRedit authorship contribution statement

Zoltán Lukács : Conceptualization, Data curation, Writing – original draft. **Tamás Kristóf** : Validation, Writing – review & editing.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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