

ELECTRICAL IMPEDANCE MEASUREMENT USING VOLTAGE/CURRENT PULSE EXCITATION

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Abstract – Electrical impedance measurements based on pulse excitation are fast and simple to implement, which makes them attractive for low-power measurement systems such as remote water conductivity monitoring based on two-electrode cells. Pulse measurements also allow us to estimate electrode parameters but measurement errors increase when the electrode resistance is very different from that of the electrolyte. We propose a dual pulse excitation technique to minimize errors in estimating electrode parameters: a voltage pulse is followed by a current pulse whose amplitude is set according to the results for the first (voltage) pulse. Experimental results for impedance networks that emulate water resistivity from 100 Ωcm to 10 $\text{k}\Omega\text{cm}$ (corresponding to surface waters) measured with two stainless-steel electrodes, yield relative errors below 0,2 % even for (emulated) corroded electrodes. For actual electrodes, relative errors are below 2 % for the resistance and 4 % for the capacitance that model its impedance. Conductivity measurements for surface waters yield relative errors about 0,1 %, which is quite acceptable for low-cost autonomous sensors. Electrode impedance estimates differ from the first to the second pulse, presumably due to the different effective current density injected by each pulse. Nevertheless, experimental results can still be used to detect corrosion in stainless steel electrodes.

Keywords: electrical impedance measurement, pulse excitation, water conductivity, electrode impedance

1. INTRODUCTION

Impedance measurement based on single pulse excitation can yield the three parameters of a Randles cell, which is a common electrical circuit model for liquid conductivity measurement based on two electrodes [1]. The measurement method is based on the circuit response to a voltage pulse lasting long enough for the resulting current through the material or measurement cell to achieve a constant value. Measuring three values of the current yields three equations whose solution is simple if the three current values are measured at carefully selected times. We select the initial and last points of the pulse and an intermediate time whose

corresponding amplitude is the average of the extreme amplitudes.

This measurement method yields an acceptable error for the liquid conductivity but the errors for the estimated electrode parameters are much higher, particularly for corroded electrodes. Sensors whose electrodes are continuously immersed would benefit from a simple method to assess electrode impedance because this impedance is related to electrode corrosion or fouling. Electrode corrosion in concrete and steel can be assessed by electrochemical impedance spectroscopy [2, 3] but this is a slow measurement method based on complex circuitry unsuited to compact autonomous systems. Time response techniques have also been implemented for both laboratory and field corrosion tests, but they normally target a single parameter: the charge-transfer resistance or the double-layer or Helmholtz's capacitance [4, 5].

We propose a double pulse injection technique to measure impedance, in which the injection of a voltage pulse is followed by a current pulse. The current measured and the impedance components estimated from the first pulse injection are used to establish the amplitude of the second pulse so that the maximal detected voltage is close to the full-scale value for the data acquisition system (DAQ). This reduces measurement uncertainty because of the finite resolution of the DAQ. This approach reduces errors in the estimated values for the components of the circuit model, at a moderate cost in complexity. Previous works that used a double/bipolar pulse technique to improve the pulse/step method only estimate the solution resistance of the conductance cell because they suppress the capacitive influence of electrodes by a time-current integration [6-8]. Here we obtain information about the electrodes, in addition to the liquid conductivity. Furthermore, using two pulses is also attractive for applications where there is no *a priori* knowledge of the amplitude of the current pulse to be applied; by first injecting a voltage pulse, we obtain a first estimate of the impedance that can be used to set the optimal amplitude and time duration for the subsequent current pulse.

2. MEASUREMENT METHOD

Fig. 1 shows the proposed measurement method, which involves two stages: (1) voltage injection – current detection and (2) current injection – voltage detection. Fig. 1a shows two elapsed pulses in voltage and current mode at times T_1 and T_2 respectively. Each pulse must last long enough (δ_1 and δ_2) until the resulting current or voltage is stable; this condition is fulfilled when the difference between adjacent samples is below the effective resolution of the data acquisition system [1]. In practice this means that C_p has to

be completely discharged before the next pulse is applied. Fig. 1b shows the equivalent electric circuit of the Randles cell (Z) [9], whose impedance is

$$Z(s) = \frac{R_s(s+1/\tau)}{s+1/\tau_p} \quad (1)$$

where $\tau = (R_p \parallel R_s)C_p$ is the time constant of the overall network and $\tau_p = R_p C_p$ is the time constant of the electrodes. Fig. 1c shows the circuit responses to the injected voltage (T_1) and current (T_2).

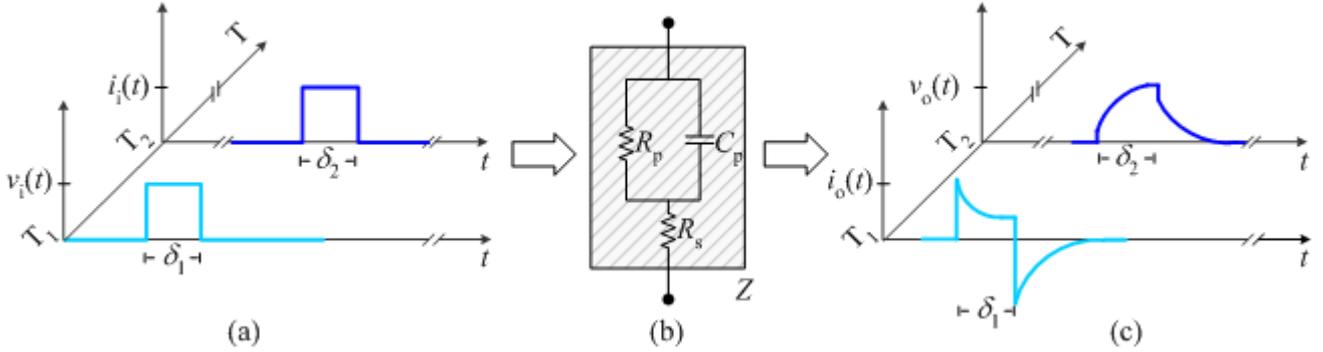


Fig. 1. Double pulse measurement method based on (a) injecting a voltage/current pulse to (b) a Randles cell and (c) measuring the current/voltage across the impedance.

When injecting a voltage pulse, the Laplace transform of the resulting current will be $I(s) = V(s)/Z(s)$ and its time expression $i_o(t)$ can be obtained from the inverse Laplace transform. The result is shown in Table 1, where V_1 is the amplitude of the input voltage pulse and $A = R_p/R_s$. From this equation, if we measure the current across the impedance at three specific times: $t_1 \cong 0$ (to obtain I_1), $t_3 \rightarrow \infty$ (to obtain I_3), and t_2 such that $I_2 = (I_1 + I_3)/2$, then a simple equation system results that leads to the expressions for R_s , R_p and C_p shown in the first row of Table 1. In practice, first I_2 is calculated and the time instant that yields the sample value closer to I_2 is assumed to be t_2 .

For the second stage (T_2), the values of the measured current I_1 and the values for R_s , R_p and C_p estimated from the voltage pulse injection are used to set the amplitude of the current pulse injected to $I_1 = \text{MSB}/(R_s + R_p)$, MSB being the most significant bit (in volts) of the data acquisition system (Fig. 2). By measuring again at $t_1 \cong 0$ (to obtain V_1), $t_3 \rightarrow \infty$

(to obtain V_3), and t_2 such that $V_2 = (V_1 + V_3)/2$, an equation system results whose solutions for R_s , R_p and C_p are shown in row 2 of Table 1.

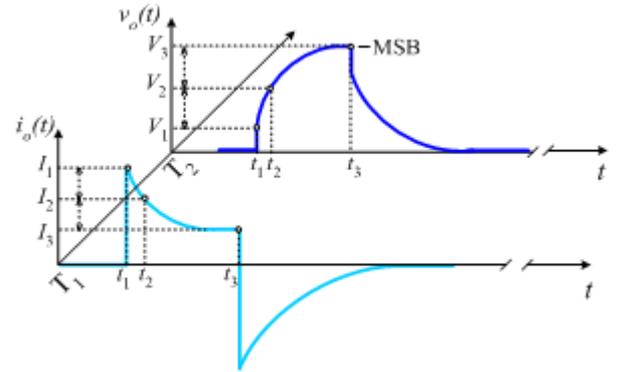


Fig. 2. Sample selection process for each pulse response.

Table 1. Parameters of a Randles cell impedance calculated from the current and voltage across the cell.

Event	Characteristic curve equation	Solution resistance	Polarization resistance	Double-layer capacitance
T_1	$i_o(t) = \frac{V_1}{R_s + R_p} (Ae^{-t/\tau} + 1)$	$R_s = \frac{V_1}{I_1}$	$R_p = \frac{V_1}{I_3} - \frac{V_1}{I_1}$	$C_p = \frac{t_2}{(\ln 2)(R_s \parallel R_p)}$
T_2	$v_o(t) = I_1 R_s \left[1 + A \left(1 - e^{-t/\tau_p} \right) \right]$	$R_s = \frac{V_1}{I_1}$	$R_p = \frac{V_3}{I_1} - \frac{V_1}{I_1}$	$C_p = \frac{t_2}{(\ln 2)(R_p)}$

3. EXPERIMENTAL TECHNIQUES

The method above has been implemented by using a data acquisition system (PXI-6221, National Instruments) as shown in Fig. 3, and applied to the measurement of impedance networks built from two precision ($\pm 0,1\%$ tolerance) metal-film resistors (R_s and R_p) and a metallized polyester-film capacitor ($\pm 5\%$ tolerance) as a grounded conductivity sensor setup. Nominal values were selected according to the target impedance (surface water conductivities measured with a stainless-steel electrode cell). From [3], the expected values for unit electrode area for stainless-steel electrodes in neutral molar solution are from $0,5\text{ k}\Omega\text{cm}^{-2}$ to $30\text{ k}\Omega\text{cm}^{-2}$, and from $0,01\text{ }\mu\text{Fcm}^{-2}$ to $40\text{ }\mu\text{Fcm}^{-2}$. Surface water resistivity is from $100\text{ }\Omega\text{cm}$ to $10\text{ k}\Omega\text{cm}$.

The PXI-6221 system was configured in triggered synchronous mode, with one analog output port (DAC with 16 bit resolution and update rate 250000 samples per second) and two analog inputs (ADC with 16 bit resolution and update rate 250000 samples per second). Triggering the system in synchronous mode reduces the uncertainty in the acquisition time and avoids the need of backward extrapolation used in [1] to obtain I_1 . The DAC controlled the amplitude of both voltage and current sources. For the voltage source (v_i), the DAC was connected to a voltage buffer working as an impedance transformer. To obtain a current source (i_i), the DAC was connected to an enhanced Howland current source based on an op amp [10]. Both op amps were AD8055, which has a low output resistance ($0,1\text{ }\Omega$), 300 MHz bandwidth and $1400\text{ V}/\mu\text{s}$ slew-rate. The excitation pulse (voltage or current) was selected by one PXI-6221 control line and an ADG884 multiplexer with low on resistance and low leakage current ($R_{on} = 0,28\text{ }\Omega$; $I_{S,D} = 0,2\text{ nA}$).

When injecting the voltage pulse, one analog input channel measured the resulting current by detecting the drop in voltage across a sense resistor [1]. The other analog input channel directly measured the voltage drop across the unknown impedance when injecting a current pulse. The system was calibrated by measuring two known R_s resistors.

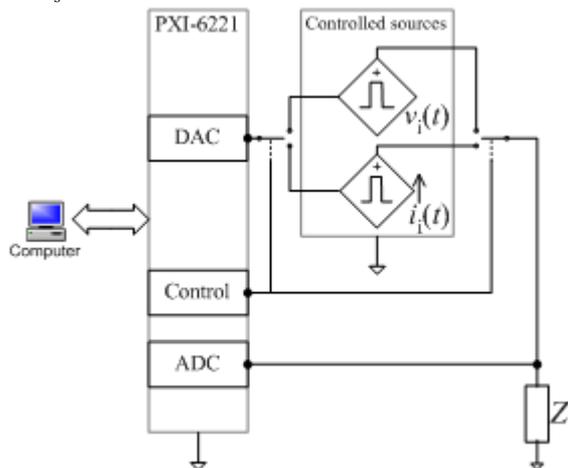


Fig. 3. Experimental setup used to assess the double pulse technique.

To implement the sensor for monitoring water conductivity based on two-electrode cells, we use a custom-built cell that uses two stainless-steel acorn nuts as electrodes and the experimental setup shown in Fig. 3. A major problem of grounded conductivity sensors is electrode polarization because of their galvanic contact with a conductive liquid. This contact stimulates electrochemical reactions at the electrode surface, which furthers corrosion processes. In order to reduce these effects, ac excitation signals are often used [11]. We have applied an alternative solution consisting of an H-bridge configuration based on analogue switches to alternate the direction of the current across the unknown impedance for successive pulses. We used the ADG884 multiplexer to implement this solution. The on resistance of the switches (R_{on}) must be subtracted from the overall series resistance measured.

4. EXPERIMENTAL RESULTS AND DISCUSSION

Fig. 4 shows the relative error of the experimental results for each of the three components of electrical networks that emulate cells with stainless-steel electrodes, one with intact electrodes and the other with “corroded electrodes”. Errors are shown as a function of the A parameter ($A = R_p/R_s$) when using the single or the double pulse technique. Corrosion studies mainly focus on a single electrode parameter, such as the polarization resistance (R_p) [4], the double-layer capacitance (C_p) [5], or the time constant of the corrosion process τ_p [12]. Here we consider the measurement of each of the two parameters that model the electrode impedance and how they affect the conductivity measurement.

Fig. 4a shows the relative error for R_s when measuring a circuit that emulates a cell with intact electrodes whereas Fig. 4b shows that error for emulated corroded electrodes (R_p value ten times smaller, as reflected in the horizontal axis). In both cases, the error for R_s is zero at the two calibration points, as expected. For both “normal” and “corroded” electrodes, a single (voltage) pulse yields a worst-case error below 1%, thus fulfilling the recommended criterion for conductivity measurements in surface water bodies [13]. The use of the second pulse further reduces the error below 0,2% in both cases. Hence, the two-pulse technique is better to measure R_s , hence water conductivity.

Similarly, Figures 4c and 4d show an error reduction for R_p , as a function of A , by more than 20% thanks to the second pulse. For the single-pulse technique, the error is larger for an “intact electrode” than for “corroded electrodes” (smaller R_p), whereas the double pulse technique yields smaller errors for the “intact electrode.” Figures 4e and 4f show that the error for C_p as a function of A reduces below half of that obtained with a single pulse. It is also observed that the error for the single-pulse technique is larger for an “intact electrode” than for “corroded electrodes” (smaller R_p), and that the double pulse technique yields smaller errors for the “intact electrode.” Errors with a single pulse are mainly due to the limited resolution in bits to detect small changes in the

measured current. Further, for the double pulse technique, errors are larger for C_p than for R_p , but the maximal relative error for the range of A values considered is below 5%, which would be quite good to assess electrode corrosion in autonomous water conductivity sensors.

The range of impedance values for stainless steel electrodes in the known bibliography is so broad that it is quite difficult to have an estimate for the electrodes of our custom-built conductivity cell. Therefore, we have used electrochemical impedance spectroscopy to determine electrode impedance by using an Agilent 4294A impedance analyzer.

We have followed the recommended practices for electrochemical impedance measurements described in [14]. First we used the impedance analyzer to measure the dummy cells (emulated electrical network) and under the same experimental conditions described for Fig. 4, over a frequency range from 40 Hz to 50 kHz, with 400 intermediate frequencies (125 Hz frequency step). Component values for the Randles cell were estimated from the frequency limits as suggested in [3]:

$$\begin{aligned} R_s &= \lim_{\omega \rightarrow \infty} |Z| \\ R_p + R_s &= \lim_{\omega \rightarrow 0} |Z| \\ C_p &= (\omega_{\max} R_p)^{-1} \end{aligned} \quad (2)$$

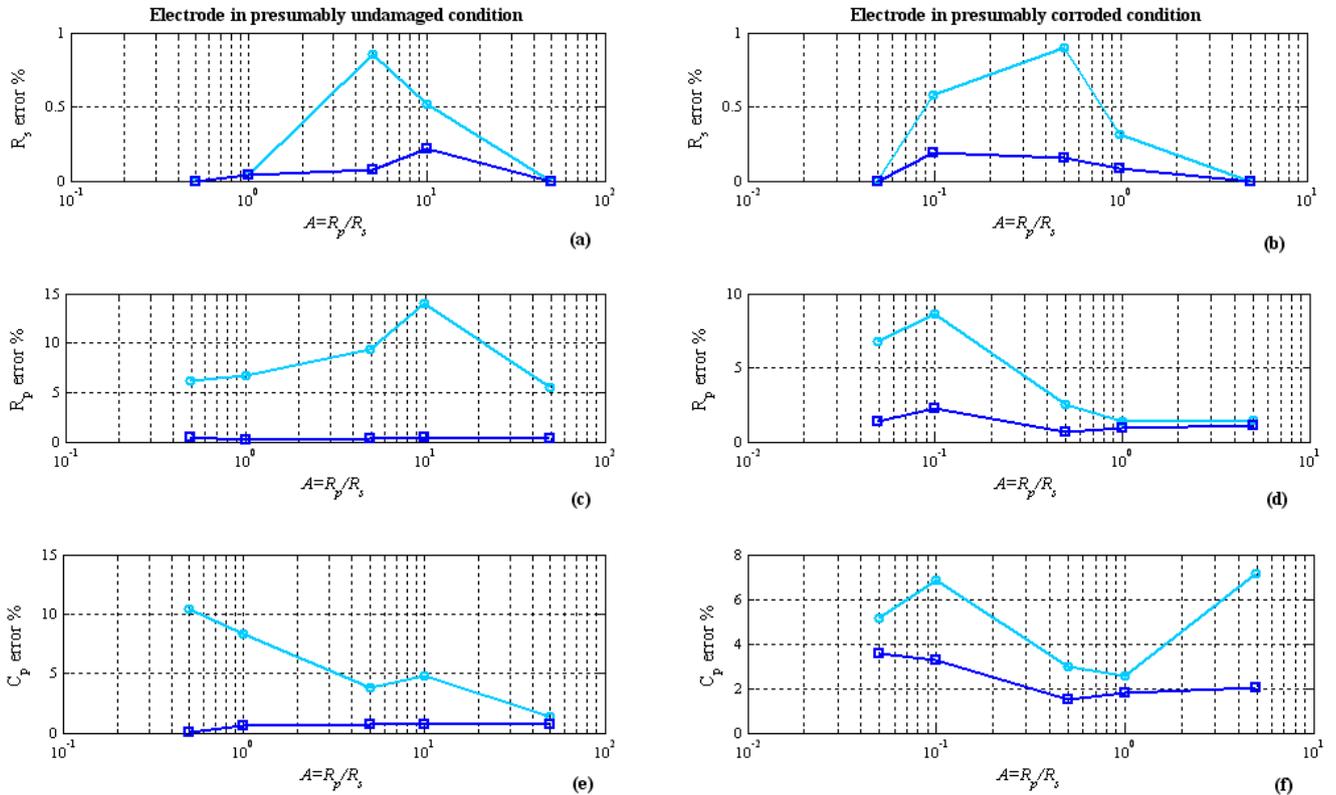


Fig. 4. Relative errors for two emulated conductivity cells in normal condition and with corroded electrodes when injecting a single pulse (light-colored line) and two pulses (darker line).

Fig. 5 shows the relative error for each component of the dummy cells in (emulated) undamaged (Fig 5a) and corroded conditions (Fig 5b), as a function of the A parameter for the same range as in Fig. 4. The largest errors are obtained for the double-layer capacitance (C_p), both for “intact” and “corroded electrodes” and are larger than those in Fig. 4 for the single- and double pulse techniques. These larger errors can be attributed to the limited frequency resolution of the impedance analyzer (125 Hz frequency step). Relative errors for the emulated charge-transfer resistance (R_p) and liquid resistivity are similar to those obtained in Fig. 4. Errors for R_p measured with the impedance analyzer can be attributed to its limited measurement capability at low frequencies, because the minimal measurement frequency is 40 Hz and we estimate the value at $\omega \rightarrow 0$ by quadratic interpolation. Similarly, relative errors for R_s can be attributed to the too low maximal measurement frequency (50 kHz), as we estimate the impedance value when $\omega \rightarrow \infty$ also by quadratic interpolation. In spite of these large errors for circuit component values estimated with the impedance analyzer, impedance components estimated for a conductivity cell can still help in assessing the results obtained with the measurement method based on a double pulse.

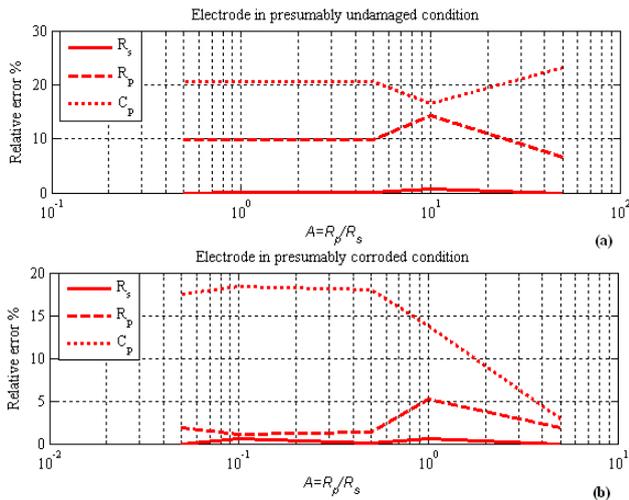


Fig. 5. Relative errors for two dummy cells in normal condition (a) and presumably corroded condition (b) obtained with the 4294A impedance analyzer.

Fig. 6 shows relative errors for the conductivity of actual surface water samples measured with the custom-built cell described above and the 4294A impedance analyzer. The system was always calibrated at two points ($100 \mu\text{Scm}^{-1}$ and 10mScm^{-1}) by using a WTW-340i conductivity meter as reference ($\pm 0,01 \text{mScm}^{-1}$ uncertainty.) Measurements were performed for the water conductivity range in Table 2.

Measurements with a single pulse excitation (light-coloured line) yielded a maximal relative error close to 1,63 %, hence below that obtained in [1], which was 5 %. The double pulse excitation technique (dark line) always yielded a smaller error than the other two techniques and reduced the maximal relative error to 0,13 %. Measurements with the 4294A impedance analyzer (dashed line) yielded a larger error than the other two techniques (maximal relative error about 7,2 %), probably because the upper measurement frequency was not high enough. Further, a Randles cell with three constant components cannot model electrochemical effects in a very broad frequency [3].

Table 2 shows the results for the electrode impedance components R_p and C_p obtained with the single and double pulse techniques as well as the 4294A impedance analyzer, for different water conductivities. All the estimated values fall inside acceptable ranges for stainless steel. Nevertheless, for a given water conductivity, there are obvious discrepancies for both R_p and C_p , which are more evident when water conductivity increases. Besides the questionable validity of the model in Fig. 1b as a possible factor to explain those discrepancies when the signal applied to the impedance being measured involves a broad range of frequencies, the different current density involved in each measurement has to be considered. Measurements based on two pulses (voltage pulse followed by a current pulse) involve a larger current density for the current pulse than for the voltage pulse, and that current density increases when water conductivity is high.

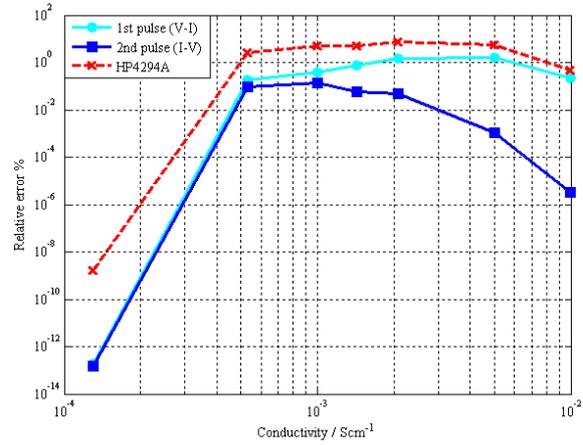


Fig. 6 Relative error for actual conductivity measurements based on dual pulse technique compared with the Agilent 4294A impedance analyzer.

Table 2 also shows a decrease in charge-transfer resistance (R_p) and an increase in double-layer capacitance (C_p) for larger water conductivities that agrees with the results reported by Geddes on the dependence of electrode models on current densities [15].

Table 2. Electrode impedance parameters.

Water conductivity WTW-340i (Scm^{-1})	V-I Technique		I-V Technique		Agilent 4294A	
	R_p ($\text{k}\Omega$)	C_p (nF)	R_p ($\text{k}\Omega$)	C_p (nF)	R_p ($\text{k}\Omega$)	C_p (nF)
130E-6	15,5	161,9	11,3	329,8	22,5	70,4
530E-6	12,9	162,6	7,4	333,8	8,51	98,3
1E-3	12,9	163,0	5,0	341,2	5,2	40,9
1,413E-3	10,7	168,5	2,6	347,2	2,3	131,5
2,09E-3	8,1	171,8	7,6	685,5	1,930	484,5
4,99E-3	6,1	184,1	2,9	708,7	0,95	1286
9,97E-3	6,0	246,7	1,9	1292	0,85	2624

Electrode diagnosis is feasible in spite of the discrepancy between electrode parameters obtained by different measurement methods. Diagnosis can be performed not only from the absolute values of electrode parameters but also from their time evolution. Therefore, given a reference condition, i.e. "intact electrodes" when they are brand new, R_p and C_p can be estimated by a given measurement technique. Next, electrode corrosion (or fouling) induced on purpose, will result in specific changes in the R_p and C_p values estimated by that technique, and these values could be used to decide whether the electrodes should be cleaned or replaced. Because the double pulse technique is more robust to the effects of R_p and C_p , it should be preferred over the single pulse technique.

5. CONCLUSIONS

The three components of a Randles cell (Fig. 1b) can be determined by injecting a voltage pulse and measuring three current values [1]. However, the finite resolution of the system implies a larger relative error for the smaller of the three currents measured, and this results in a larger relative error for the two components that model the electrode impedance than for the series element, which models water conductivity. Injecting a current pulse and measuring three voltages also allows us to determine the three impedance components. Furthermore, if first a voltage pulse is injected and the results are used to adjust the intensity of a subsequent current pulse, the relative errors for the three components of the cell are smaller than when using a single pulse. Experimental results for a cell emulated by an electrical network whose series resistance emulates surface water conductivity yield relative errors below 0,2 % for R_s and below 5 % for R_p and C_p . For actual water measurements performed with a custom-built conductivity cell that uses stainless-steel electrodes, the error is 0,13 %, well below of the 5 % limit set in water quality standards [13].

Estimated values for electrode components R_p and C_p differ from the single pulse to the double pulse technique. Impedance spectroscopy performed with a commercial impedance analyzer (Agilent 4294A) is not of much help as a Randles cell with constant components cannot describe the electrochemical phenomena in a conductivity cell in a broad frequency range. A possible explanation for the dependence of R_p and C_p on the measurement technique, as shown in Table 2, is the different current densities involved in each measurement method. The decrease of R_p and the increase of C_p for increasing water conductivities, as observed in Table 2, agree with results reported in [14]. In any case, the double pulse technique detects changes in R_p and C_p , in addition to accurately measure R_s , and therefore it can be used to diagnose the state of the electrodes if first data about R_p and C_p for corroded electrodes are obtained with the same measurement method.

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