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Electrical impedance measurement using pulse excitation

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Abstract- Impedance is the only electrical property common to all materials. Its measurement is used for material characterization and testing, and for sensor design. Impedance measurements in a broad frequency range are slow and need complex instrumentation. Impedance measurements in the time domain can be faster but need more computation power. We propose a novel measurement method intended for applications where the unknown impedance can be modeled by three independent components. The method consists of applying a single pulse, measuring three amplitudes, and calculating the unknown components from the analytical solution of the resulting equation system. The three measurement points are so selected that the computation power needed is minimal. Furthermore, measurements at the leading edge of the pulse are avoided by using linear extrapolation. The method suits low-power measurements in electrolytes, which involve electrodes that can be modeled by a resistance shunted by a capacitance. Actual conductivity measurements from 100 µS/cm to 10 mS/cm, yield relative errors below 3.5 %, which fulfills the 5 % target set in some standards for monitoring the conductivity of surface water bodies.

I. Introduction

Electric impedance measurements are used for characterizing materials, electronic circuits and components, for sensing, and for interpreting physicochemical phenomena. Impedance is calculated from the direct relation between electrical currents and potential differences at a given frequency. In chemical measurements, impedance is proportional to concentrations, activities or partial pressures of chemical species [1].

Impedance is defined in the frequency domain but it can be measured in both the frequency and time domains. Depending on the impedance range, the relative value of their real and imaginary parts, desired resolution, parasitic impedances involved and property to be analyzed, there is a variety of measurement methods with different trade-offs. Impedance spectroscopy (IS) involves the injection of sine voltages (or currents) in a broad frequency range and detecting the resulting current (respectively voltage drop) in the material being analyzed [2]. This is a slow method which requires the synthesis of sine signals with high spectral purity and coherent amplitude demodulation for best signal-to-noise ratio. On the other hand, time-domain measurements using a step signal or pulses benefit from simple stimulus generation but need fast analog-to-digital converters (ADC) and enough data points to reconstruct the input-output frequency response by means of the Digital Fourier Transform [3]. Digital interpolation techniques can improve resolution, but the method needs some computation power.

Model-based impedance measurements can benefit from the *a priori* knowledge about the impedance being measured to simplify instrumentation design and reduce measurement time. This is the case, for example, in electrochemical impedance measurements involving electrolytes, such as conductivity measurements, which need electrodes to translate ion currents in the electrolyte into electron currents in the electronic circuit. In a first-approach analysis, electrodes can be modeled by a resistance shunted by a capacitance, and the electrolyte can be modeled by a series resistor [1]. Also, some passive electronic components can be modeled by an ideal passive component (R, L, C) plus some series or parallel components that describe parasitic effects.

Three independent impedance components can be determined by using a square wave voltage and measuring current at three specific times [4]. Solving a system of three equations yields each of the impedance components. Here we propose a simpler technique based on a single pulse excitation and three amplitude measurements in the response signal. The theoretical response is easier to obtain than that

resulting from the excitation with a periodic signal, and the computing load to calculate the unknown components is lighter, which makes the system attractive for low-power measurement systems.

II. Measurement method

Figure 1 shows the proposed measurement method. Figure 1b is a simplified Randles cell, which is an electrical equivalent circuit commonly used for electrochemical impedance measurements and other applications [5-8]. This model includes a solution resistance R_w plus any series resistance of the electrodes (R_{se}), that is, $R_s = R_w + R_{se} + R'_{se}$, a double layer capacitance due to the charge transfer process (C_p) at the electrode-electrolyte interface, and a charge transfer or polarization resistance (R_p) [1]. We are particularly interested in water quality estimation from measurements performed with two-electrode conductivity cells. The impedance of a Randles cell is

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

where $\tau = (R_p \,||\, R_s)C_p$ is the time constant of the overall network and $\tau_p = R_pC_p$ is the time constant of the electrodes. If a single voltage pulse $u(t)$ lasting a time δ is applied across that impedance (figure 1a), the resulting current (figure 1c) is $I(s) = U(s)/Z(s)$. If we measure that current during δ, we need to analyze only the response to the first voltage transition (voltage step). The inverse Laplace transform of $I(s)$ is then

$$i(t) = I_F\left(Ae^{-t/\tau} + 1\right) \tag{2}$$

where $A = R_p/R_s$ and I_F is the direct current through the cell, also called Faradic current, defined as,

$$I_F = \frac{U_0}{R_s + R_p} \tag{3}$$

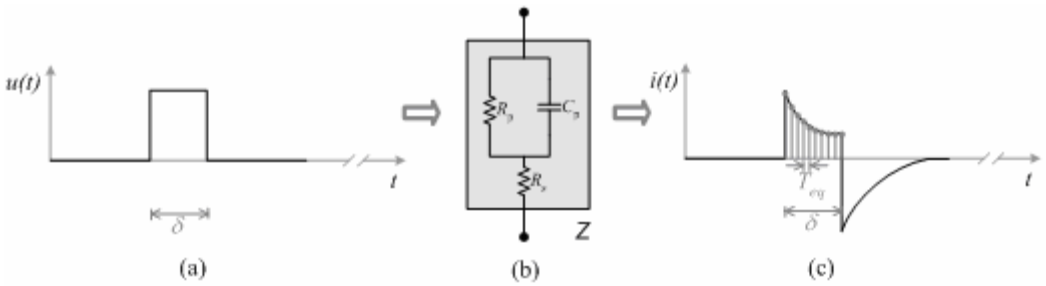

Figure 1. Impedance measurement by (a) applying a voltage pulse to (b) a Randles cell and (c) measuring the current across the impedance.

We propose to measure $i(t)$ at three given times t_1, t_2 and t_3, and solve the three-equation system. Since (2) involves an exponential function, solving the equation system for three arbitrary times may need calculation power beyond the capabilities of low-power compact systems. However, by selecting $t_1 \cong 0$, $t_3 \to \infty$ and t_2 arbitrary, we obtain $i(t_1) = I_1 = I_F(1 + A)$, $i(t_3) = I_3 = I_F$, and $i(t_2) = I_2$ as given by (2). These three equations lead to

$$\begin{aligned} R_s &= \frac{U_0}{I_1} \\ R_p &= \frac{U_0}{I_3} - \frac{U_0}{I_1} \end{aligned} \tag{4}$$

To obtain C_p, we select I_2 to maximize the difference between the measured currents, $I_1 - I_2 = I_2 - I_3$,

$$C_p = \frac{t_2}{(\ln 2)\left(R_s \,||\, R_p\right)} = \frac{t_2}{(0.693)\left(R_s \,||\, R_p\right)} \tag{5}$$

The time t_2 is estimated by linear interpolation between those corresponding to the sampled current values

closer to I_2. Time t_1 is that corresponding to the first valid data sample and t_3 belongs to the flat zone for the resulting current curve (figure 1c). Because measuring at the trailing edge of the pulse can result in gross errors, we calculate the value at t_1 by backward linear extrapolation from the two next samples [9]. We estimate t_3 by looking for adjacent current values whose difference is below the effective resolution of the data acquisition system. That is,

$$\left|I_3 - I_\infty\right| \le \mathrm{LSB}/2^M \tag{6}$$

where M is selected according to the required accuracy and LSB is the least significant bit defined as

$$1\ \mathrm{LSB} = \frac{I_F\left(A+1\right)}{2^N} \tag{7}$$

N being the number of bits of the ADC in the data acquisition system. Solving (6) leads to

$$t_3 \ge \left[\left(M+N\right)\ln 2 - \ln\left(A+1\right) + \ln A\right]\tau \tag{8}$$

which implies either a previous knowledge of the order of magnitude for the impedance components to be determined, hence τ, or an iterative measurement process involving different durations for the voltage pulse.

III. Experimental techniques

The proposed technique has been first validated by computer simulation using Matlab® to roughly assess the effect of measurement uncertainty propagation for different values of the unknown impedance elements, and also the effects of the uncertainty in actual measurement times. Theoretically, the method works right for any triad of measurement times, but the actual amplitude and time resolution of any measurement system will be limited, particularly in low-power systems.

We built the measurement setup in figure 2 and applied it to an impedance network consisting of two precision (±0.1 %) metal-film resistors (R_s and R_p), and a metallized polyester-film capacitor (±5 % tolerance). R_m is the current-sensing current resistor. Component values were selected according to the target impedance, namely water conductivity measurement for environmental monitoring of surface water bodies (from drinking water to waste water) based on a two-pole cell arrangement. This implies conductivities from 100 µS/cm to about 10 mS/cm, hence resistivities from 100 Ω cm to 10 kΩ cm that will determine R_s depending on the geometry and separation between electrodes. Electrode size, material and fouling effects result in a broad range of possible values for R_p and C_p. According to [10], for stainless steel electrodes in a neutral molar solution, the expected values per unit area are from 500 Ω cm^{-2} to 30 kΩ cm^{-2}, and from 0.1 µFcm^{-2} to 40 µFcm^{-2}. Actual resistor values in our test network were measured by a digital multimeter (Agilent 34401A, relative uncertainty about ±0.002 %), and capacitor values were measured by an impedance analyzer (Agilent 4294, relative uncertainty below 1 % at 1 kHz and 0.5 V oscillator level). Input pulse voltages (1 V) and adjustable δ time were provided by a function generator (Agilent 33220A) whose typical relative uncertainty for this range is ±1 mV and 300 ps plus 10^{-7} of signal period, and pulse rise time below 13 ns. The range for δ for the emulated conductivity cells is from 8 µs to 300 ms. The voltage buffer (AD8055) is an impedance transformer to obtain less than 0.1 Ω at 1 Hz and 2 Ω at 10 MHz, well below the 50 Ω output resistance of the function generator.

The drop in R_m was measured with a 16 bit data-acquisition card (PXI 6221, National Instruments) that obtained 250,000 samples each second. For the duration of the voltage pulse applied, this means an uncertainty well below 4 µs for the values of the measurement times (t_1, t_2, t_3). The experiment was repeated 50 times and the average of the 50 values of R_s, R_p and C_p was calculated.

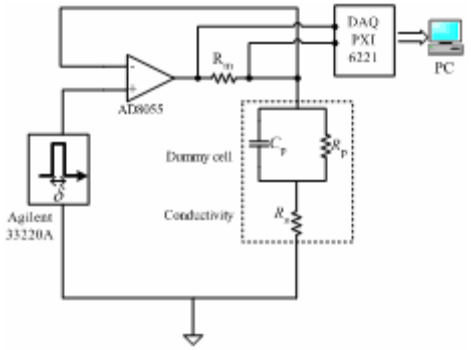

Figure 2. Equivalent circuit for the measurement method implemented.

IV. Experimental results and discussion

Figure 3 shows the relative error of experimental results for each of the three components of nine networks that emulate several cells with stainless steel electrodes immersed in surface water bodies. Measurements were calibrated by using the end-range values for R_s (100 Ω and 10 kΩ) and an electrode impedance 1 kΩ||4.7 μF, which are typical values [10]. The maximal relative error obtained is about 1.6 % for R_s and it happens for the largest C_p (figure 3a). If instead of estimating the amplitude at t_1 by backwards extrapolation, we use the amplitude of the first sample measured, the maximal relative error for R_s increases to 19 %.

The relative error for R_p increases with R_s (figure 3b) and is larger for the electrode with the lowest resistance, hence closer to R_s in the measurement range tested. Its maximal value is about 15 %. The relative error for C_p increases for large and small values for R_s and also depends on R_p (figure 3c). Its maximal value in the range tested is also about 15 %. Low-impedance networks have a short time constant and yield a fast transient response that is quite difficult to measure. Hence, severely corroded electrodes should display this effect, which offers a method to detect their status provided the resolution and sampling speed of the data acquisition system are high enough. Amplitude resolution affects the accuracy in the three calculated components, whereas sampling speed affects only R_s and C_p. Less corroded electrodes should result in intermediate time constants, hence impedance components easier to measure.

Actual water conductivity measurements using a custom-built conductivity cell based on two stainless-steel electrodes were performed by the same instrumentation in figure 2. Four electrolyte samples were measured, whose actual conductivity was determined by a WTW-340i conductivity meter (± 1 digit uncertainty in each of its five measurement ranges). Two-point calibration was applied by using the minimal and maximal conductivities as reference values. We also calculated the cell constant from the measured R_s value and the actual conductivity for the smaller conductivity value in our range. The maximal relative error for the four conductivities measured was 3.5 %, which is smaller than the recommended 5 % target for continual on-line monitoring water monitoring systems [11]. It must be borne in mind that R_s includes the electrolyte resistance and any series resistance because of the electrodes. Therefore, any variation in electrode series resistance with solution concentration will not be compensated for. Rather, it will result in an apparent variation of the cell factor with concentration.

We have also calculated the electrode impedance for our custom-built cell. The results were from 1996 Ω to 3100 Ω for R_p and from 2.09 μF to 490 nF for C_p. These values are inside the electrochemical range for stainless steel [10]. However, we expected them to be constant regardless of the concentration. Therefore, further analysis is needed to determine if those variations were a result of uncertainty propagation, of the equivalent model for the electrodes was too simple to describe all interface processes in them.

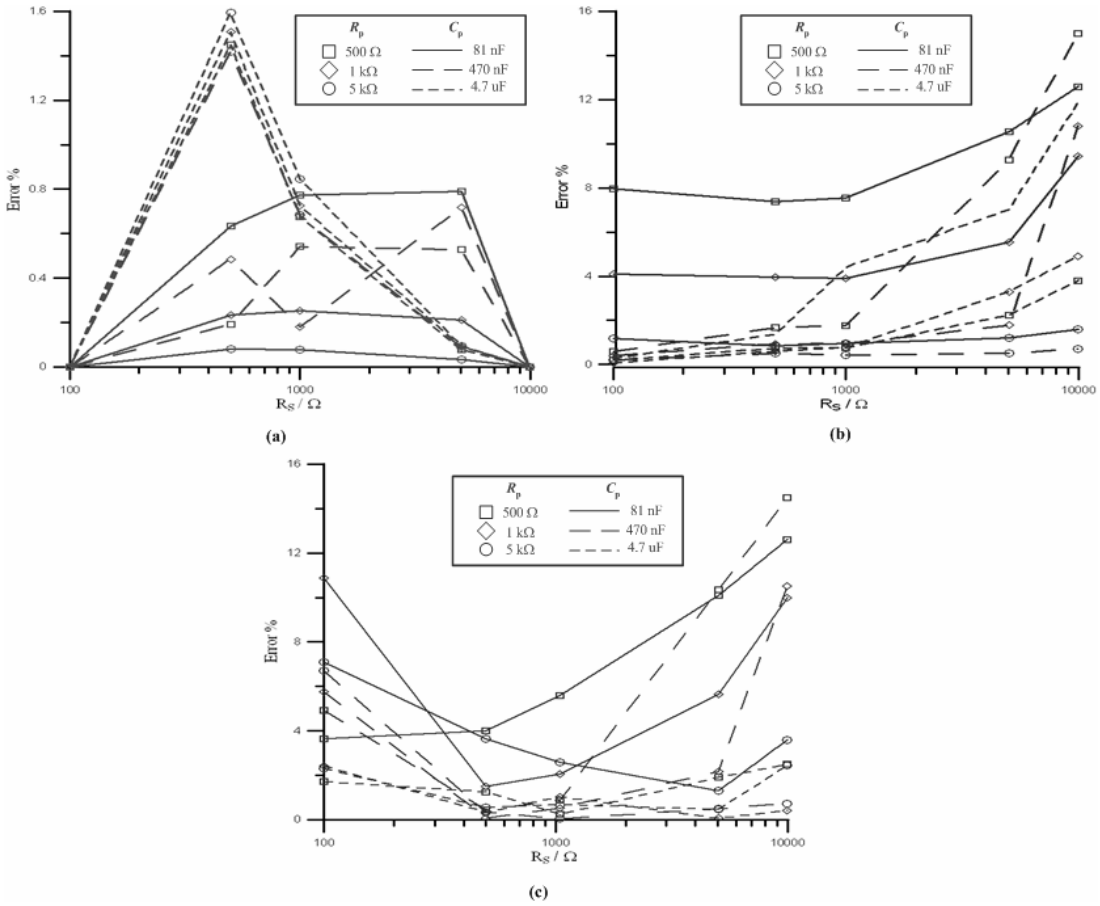

Figure 3. Relative errors for several electrical networks that emulate conductivity cells with different solution conductivities and electrodes. (a) Series resistance, R_s; (b) Electrode polarization resistance, R_p; (c) Electrode double-layer capacitance (C_p).

V. Conclusions

We propose a measurement method to identify three independent components of an impedance network, such as those used to model several electrochemical phenomena, for example water conductivity measurements based on two electrodes. The method uses a single pulse excitation and measures three amplitudes of the response signal at three given time instants, so selected as to minimize the computation power needed to solve the resulting equation system. The selected times are the initial point, a point far enough for the response to be settled around its final value, and an intermediate point such that the amplitude response is the average of the initial and final amplitudes. Equations (4) to (6) allow then the calculation of the three impedance components using only simple arithmetic operations. To improve the time resolution, the time for the intermediate point is estimated by linear interpolation between the two samples whose values are closer to the average of the initial and final amplitudes. To avoid gross errors resulting from transients due to the trailing edge of the pulse, instead of the value of the first sample acquired, we estimate that value by backwards linear extrapolation from the second and third samples. This method suits low-power, compact equipment, such as that required for monitoring the conductivity of surface water bodies using two-pole cell arrangements.

Experimental measurements of passive networks that model such conductivity cells, using a 16 bit system that acquires 250,000 samples each second, yield relative errors below 1.6 % for the emulated conductivity (from 100 µS/cm to 10 mS/cm) and 15 % maximum for the parallel resistance and capacitance that model the electrodes. Actual water conductivity measurements in that range yielded maximum 3.5%, which is less than the recommended error for monitoring surface water bodies [11]. However, estimates for electrode impedances depended on the conductivity, which should not be case if the method should be able to detect electrode corrosion, fouling or aging. Further work is needed to identify uncertainty sources and their propagation.

Acknowledgments

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