Examining and modelling the influence of lengths of rebars in concrete to shapes of impedance spectra

1. Introduction

In the advanced corrosion diagnostics of reinforced concrete structures there have been recent attempts to apply the electrochemical impedance spectroscopy method. For reinforced concrete elements EIS tests are done using the potentiostat 1 in the three-electrode system (Fig. 1), in which the working electrode 2 is the steel reinforcement, while the counter electrode 3 with the reference electrode 4 are placed in the measuring head 5 on the concrete surface. The most difficult problem of EIS tests performed on large reinforced concrete elements is the correct identification of the range of polarization 6 of rebars. The problem results from difficulties in testing steel rods completely sheathed with concrete, as well as the influence of heterogeneity in concrete structures on paths of conducting the alternating current - see (1). In the case of impedance tests the polarization areas of steel in concrete can be identified by limiting the distribution of the electric field by means of an additional counter electrode - see (2). This is not, however, fully effective.

Another problem of implementation of the EIS test method for reinforced concrete structures is the considerable lengths of the reinforcement, that is, large areas of the working electrode in comparison to conventional laboratory tests on small samples of metal – see Fig. 1a. With the possibility of polarization during EIS tests on such a large steel surface, the question arises of the influence of reinforcing rod length on the obtained impedance spectra



Fig. 1. The idea of impedance tests on the reinforcement of the reinforced concrete structures: a) view of the measuring system on a part of the reinforced concrete beam, b) the problem of identifying the polarization surface of long reinforcing bar in concrete

shapes. This problem is mentioned indirectly in (2), in which by analysing long reinforcing rods there is proposed a model using a steel-concrete system composed of the Randles circuit connected in parallel. From the experimental side in the publication (3) presenting the impedance test on large reinforced concrete slabs it was shown that with an increasing number of polarized bars the circuit impedance decreases. This paper attempts an experimental-analytical evaluation of the influence of the length of rebar in concrete on the shape of impedance spectra obtained by diversified areas of counter electrodes.

2. Course of experimental tests

Experimental studies were performed on two series of concrete test elements with a single reinforcing bar of smooth steel grade S235JR. In both series, the composition of the concrete mix per 1 m³ of concrete was: 489 kg of Portland cement 32.5 R, 501 kg of fine aggregates to 2 mm, 1168 kg of coarse aggregate fraction of 2-8 mm and 212 liters of water. In one series of samples 2% CaCl₂ by weight of cement was added to the concrete mix, to induce corrosion of the reinforcement in the concrete. In the second series of samples no such additions were applied to cause passivation of reinforcing steel.

Each of the two series consisted of five rectangular elements of equal cross-sections of 100×100 mm and varying lengths of 50, 100, 150, 200 and 250 mm – Fig. 2. In each sample a single rein-

forcing bar diameter of 16 mm was located in the longitudinal direction and led out one side of the 30 mm of concrete. Minimum concrete cover of reinforcement in each specimen was identical at 20 mm.

Impedance studies of the reinforcement of concrete elements were conducted in a test station as shown in Fig. 3. Measurements were taken in the three-electrode system with the working electrode 1 in each of the five samples being a reinforcing bar of the same diameter and different lengths of 50, 100, 150,



Fig. 2. The test elements of one series



Fig. 3. Stand for impedance tests of the reinforcement of concrete specimens: 1 – working electrode, 2 – reference electrode, 3 – counter electrode, 4 – foil, 5 – felt, 6 – concrete ballast, 7 – potentiostat

200 and 250 mm. Reference electrode 2 was an electrode Ag|AgCl in a plastic housing. However, as counter electrode 3 stainless steel was used. All sheets have the same thickness of 2 mm and a width of 100 mm, while the length corresponded to the longitudinal dimensions of the samples. In each the counter electrode 3



Fig. 4. Summary of the Nyquist plot of impedance spectra for different lengths of reinforcement in concrete: a) without additives, b) with the addition of 2% CaCl₂

in the geometric centre of gravity was made by a hole diameter of 7 mm, allowing the introduction of the reference electrode 2.

In order to stimulate electrode processes the concrete samples were immersed in tap water to half the depth of the reinforcement for 24 hours before the impedance measurements. To minimize the impact of changes in humidity of the concrete during the measurement, test elements were protected with

foil. Each counter electrode 3 was placed on the upper surface of the concrete sample by wet felts 5, to ensure proper electrical contact of the counter electrode and the reference electrode with the concrete. Uniform adherence of the counter electrode through the concrete felt was forced by the concrete ballast 6 ensuring a clamp force of 30 g/cm².

The impedance tests were performed by a Gamry Reference 600 potentiostat 7, after about 2-3 hours of potential stabilization. Measurements carried out in potentiostatic mode, with fixed frequencies of 1 MHz to 10 mHz and amplitude of the potential of 10 mV relative to the stationary potential of the reinforcement.

3. Results of the impedance tests

Fig. 4 shows on a complex plane the results of impedance tests on reinforcement of varying lengths located in concrete without additives (Fig. 4a) and in concrete containing chlorides (Fig. 4b).

In spite of the realization of measurements on various test elements, in which there is no possibility of obtaining the same electrochemical properties of concrete and steel, the results show a very characteristic tendency. The greatest impedance of the steel-concrete

> system was characterized by the smallest test element whose the length of reinforcing bar was 50 mm. The lowest impedance showed the largest sample of concrete with reinforcement 250 mm long. In the case of reinforcing rods in concrete without additives the shape of impedance spectra (Fig. 4a) was very typical for the passive steel – see (4). However, in the samples of concrete containing chlorides on the Nyquist plot (Fig. 4b) in the low frequency range the steel was characterized by two time constants, and the shape of the distribution of points may indicate the course of corrosion processes.

> In order to carry out the quantitative assessment obtained in Fig. 4a and 4b test results there was used an electrically equivalent circuit taken from (4), shown in Fig. 5c. In the circuit diagram modelling the steel-concrete system, R_1 designates the resistance of the concrete pore liquid, while R_2 and R_{2a} , together with CPE_2 and CPE_{2a} , characterize respective-

ly the resistance and capacitance of the electrical double layer formed at contact in the liquid phase and solid phase of concrete. A more detailed explanation of the physical meaning of the parameters R_1 , R_2 and R_{2a} and CPE_2 and CPE_{2a} , based on the so-called alternating current conductive paths in concrete, can be found in (1). In addition to the electrochemical characteristics of concrete in the circuit in Fig. 5c, there is an element Rt with CPE characterizing an appropriate charge transfer resistance, and a double layer capacitance at the interface of the metal-concrete pore liquid. The fourth of the constant phase elements, CPE₃, in the discussed model characterizes the properties of the transitional zone between the steel and concrete.

Table 1

THE RESULTS OF IMPEDANCE SPECTRA ANALYSIS – CHARACTERISTICS OF CONCRETE

concrete	steel rod	R_1	R ₂	$R_{\rm 2a}$			CPE _{2a}	
	L				Y ₂	α ₂	Y_{2a}	$lpha_{2a}$
	mm	Ω	Ω	Ω	pF s ^{α-1}		pF s ^{α−1}	
without additives	250	0.101	223	285	694	0.978	347700	0.639
	200	1.176	112	765	667	1.000	15570	0.800
	150	0.029	1336	908	6269	0.926	2013000	0.491
	100	0.032	2275	557	7462	0.808	150800	0.764
	50	0.230	5218	3554	1290	0.983	744400	0.488
with chlorides	250	0.001	0.010	294	6272	0.648	8933	0.832
	200	0.001	0.003	0.03	710	0.508	0.312	0.550
	150	0.001	0.003	688	5626	0.867	6013	0.841
	100	0.010	0.013	731	3776000	0.775	3097	0.864
	50	0.002	0.002	1402	1153	0.875	435	0.936

Table 2

THE RESULTS OF IMPEDANCE SPECTRA ANALYSIS – CHARACTE-RISTICS OF STEEL

ete	steel rod	CF	PE ₃	CF	P	
ncre	L	Y ₃	α3	Y ₀	α	Γ, r
8	mm	μF s ^{α-1}		μF s ^{α-1}		kΩ
without additives	250	11270	0.682	3915	0.547	0.202
	200	17980	0.886	2323	0.537	1.146
	150	6817	0.780	2795	0.599	0.588
	100	4458	0.697	1321	0.290	3.909
	50	2061	0.676	1095	0.510	6.248
	250	21230	0.320	3.76	0.624	0.123
es	200	5484	0.163	0.001	1.000	0.411
with Iorid	150	14830	0.213	10.24	0.484	0.494
с Ч	100	113300	0.768	44.57	0.409	0.581
	50	3815	0.273	25.53	0.441	0.803

In Tables 1 and 2 are presented the results of the 10-th impedance spectra shown in Fig. 4a and 4b. Table 1 presents the characteris-

tics of the concrete, while table 2, the characteristics of the reinforcing steel. The numerical values of all the electrochemical parameters were determined by the iterative fitting algorithms of the Downhill Simplex method, using the application Gamry Echem Analyst[™].

4. Model of steel-concrete system

In order to make an attempt to explain the observed phenomena in Fig. 4a and 4b, i.e., the tendency of impedance spectra to change shape as a function of changes in length of reinforcing rod and the counter electrode surface, the steel-concrete system model shown in Fig. 5b was proposed.

The model characterizes any length *L* of reinforcing rod 1 of ϕ diameter placed in the concrete element 2 – see Fig. 5a. The minimum thickness of reinforcement cover in the direction of a counter electrode 3 equals *c*. The counter electrode 3, together with the reference electrode 4 placed on the surface of the concrete form a three-electrode system connected to a potentiostat 5.

A reinforcing rod of *L* length is divided into 2*n* short sections 6 of the same length ΔL , arranged symmetrically about the axis of the counter electrode 3 and the reference electrode 4, hence $L = 2n \times \Delta L$. Each vertical strip of concrete separating the width ΔL together with a fragment of the reinforcing rod with an area of $\Delta S = \pi \phi \times \Delta L$ describes the Z_{ci} impedance characterizing concrete in series with the Z_{si} impedance characterizing steel.

It is assumed that the L_{CE} length of the counter electrode is equal to the *L* length of the reinforcement and that the I_p range of the polarization during the impedance measurement comprises the entire reinforcement, i.e., $I_p = L$. Current lines 7 resulting in the concrete between the counter electrode 3 and the polarized surface of the reinforcement 8 are straight segments [see (2)] of length $I_i = c + \phi/2$.



Fig. 5. Model of steel-concrete system to simulate the influence of the length of reinforcing bar in concrete and the area of the counter electrode on the shape of impedance spectra

A so-called elementary electrical equivalent circuit (Fig. 5c), composed of elements electrochemically characterizing concrete and steel, and optimally matched to the results of impedance tests is introduced to the model. In the context of the test results shown in Figs. 4a and 4b the described requirements well meet the circuit illustrated in Fig. 5c. Taking into account the circuit shown in Fig. 5c, there are introduced to the model of the steel-concrete system shown in Fig. 5b electrochemical characteristics of steel (with no α -parameters), calculated proportionally to the elementary polarization areas ΔS of the reinforcement – see (5), i.e.,

$$R_{ti} = \frac{R_t}{\Delta S}, \quad Y_{0i} = Y_0 \cdot \Delta S, \quad \alpha_i = \alpha,$$
$$Y_{3i} = Y_3 \cdot \Delta S, \quad \alpha_{i3} = \alpha_3.$$
[1]

At the same time the electrochemical characteristics of concrete (also with no α -parameters) are introduced taking into account the scale factor – see (5), i.e.,

$$R_{1i} = R_1 \frac{I_i}{\Delta S}, \quad R_{2i} = R_2 \frac{I_i}{\Delta S}, \quad Y_{2i} = Y_2 \frac{\Delta S}{I_i},$$
$$\alpha_{i2} = \alpha_2, \quad Y_{2ai} = Y_{2a} \frac{\Delta S}{I_i}, \text{ etc.}$$
[2]

In relations [1] parameters without the *i*-letter in subscript (e.g., R_t , Y_0 , etc.) characterize the electrochemical properties with respect to a unit area of the reinforcing steel. But in relations [2] the same parameters (e.g., R_1 , R_2 , Y_2 , etc.) describe the electrochemical properties with respect to unit volume of concrete. Finally, the total equivalent impedance of the modelled steel-concrete system (Fig. 5b) can be determined from the expression:

$$\frac{1}{Z} = \sum_{i=1}^{n} \left(\frac{1}{Z_{ci} + Z_{si}} \right),$$
[3]

where

 $Z_{ci} = Z_{ci} \left(R_{1i}, R_{2i}, R_{2ai}, Y_{2i}, \alpha_2, Y_{2ai}, \alpha_{2ai} \right)$ and $Z_{si} = Z_{si} \left(R_i, Y_{0i}, \alpha, Y_{3i}, \alpha_{3i} \right).$

5. Simulation of the impedance spectra of rebars

Based on the assumptions of the steel-concrete system model described above (Fig. 5) a computer application was developed to simulate the influence of variable length of reinforcement and variable areas of counter electrode on the shapes of the obtained impedance spectra. The numerical simulation adopted as a starting point the electrochemical characteristics of two reinforced specimens with lengths L = 50 mm (one made of concrete without additives, the second with the addition of chloride). For these two specimens the appropriate parameters listed in Tables 1 and 2 were introduced into the model, after calculations in accordance with the transformed relationships [1] and [2]. Assuming I_i = 2.8 cm and $\Delta S = \pi \times 1.6 \times 5 = 25.12 \text{ cm}^2$, then for example, for specimens without additives the value introduced was $R_2 = 5218 \times (25.12/2.8)$ = 46813 Ω . Under these assumptions distributions of the model spectra were generated which characterize the other tested concrete specimens with rebar lengths of *L* = 100, 150, 200 and 250 mm – see Fig. 6.

The model simulation of the distribution of impedance spectra shown in Fig. 6a and 6b gave a similar image to the empirical distributions shown in Fig. 4a and 4b. It should be noted that full agreement of the experimental distributions with the model distributions is impossible because of the independent production and ripening of concrete test elements. Furthermore, the proposed method of simulation of alternating current polarization for any length of reinforcement is automatically suited to simulate, though in practice difficult to identify, any polarization range in long reinforcements in concrete. The described possibility can be used in studies of corrosion of reinforced concrete.

6. Summary

Information obtained from experimental testing show a distinct change in the impedance spectra shapes of the reinforcement as a function of its length (Fig. 4). The testing results explained by the described model of a steel-concrete system (Fig. 5) may be useful in the diagnosis of corrosion of reinforced concrete to formulate new methods which identify the ranges of polarization of reinforcement in concrete.

The article was developed as part of the Project No. POIG. 01.01.02-10-106/09-00 of Innovative Economy Operational Programme.



Fig. 6. Model simulation of concrete reinforcement impedance spectra distribution in concrete: a) without additive, b) with chlorides

References

1. G. Song, Equivalent circuit model for AC electrochemical impedance spectroscopy of concrete, Cem. Concr. Res., **30**, 1723 (2000).

2. L. Lemoine, F. Wenger, J. Galland, Study of the Corrosion of Concrete Reinforcement by Electrochemical Impedance Measurement, Corrosion Rates of Steel in Concrete, p. 118, ASTM STP 1065, Philadelphia1990.

3. M. F. Montemor, A. M. Simoes, M. G. S. Ferreira, Chloride-induced corrosion on reinforcing steel: from the fundamentals to the monitoring techniques, Cem. Concr. Comp., **25**, 491 (2003).

4. C. A. Scuderi, T. O. Mason, H. M. Jennings, Impedance Spectra of Hydrating Cement Pastes, Journal of Material Science **26**, 349 (1991).

5. S. J. Ford, J. D. Shane, T. O. Manson, Assignment of features and Impedance Spectra of the Cement-Paste/Steel System, Cem. Concr. Res., **28**, 12, 1737 (1998).