Effects of a Constant Magnetic Field on the Electrochemical Reactions of Quercetin

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The paper presents a study of the effect of a constant magnetic field (CMF) on the basic processes of quercetin electrochemical reactions. According to the observation made in previous studies, the presence of a double bond in the C-ring of quercetin enhances the antioxidant properties of that compound, whereas the presence of –OH groups also affects the antioxidant properties. Using cyclic voltammetry it was found that the constant magnetic field improves the efficiency of quercetin electrooxidation, especially of the third stage of the process, i.e. the stage in which the oxidation of the OH groups in the A-ring is the most difficult. The use of HPLC confirmed the electrochemical measurements and the results of cyclic voltammetry studies. The beneficial effect of the magnetic field on the efficiency of quercetin oxidation was confirmed by the results of impedance spectroscopy measurements.

1. Introduction

Quercetin belongs to a group of flavonoids that are present in fruit, vegetables and wine. The presence of a double bond in the C-ring enhances the antioxidant properties of quercetin. The presence of –OH groups at positions 3,5,7,3′,4′ not only affects the antioxidant properties, but also increases the chelation capacity of divalent metal cations, which catalyze the oxidation processes, thus affecting significantly the inhibition of oxidation processes, which are not always beneficial to the cells.[1–3] Quercetin demonstrates potent anti-inflammatory properties, prevents cardiovascular diseases and inhibits the proliferation of tumor cells.

The quercetin electrooxidation process, for example on the surface of the glassy carbon electrode, occurred in three stages. The oxidation of catechol OH groups in the B-ring is the easiest (Figure 1)(within the lowest range of potentials – the highest antioxidant activity). Then, the OH group in the C-ring is oxidized. The oxidation process of the OH groups in the A-ring occurs at the highest potentials.

A. M. O. Brett et al. studied the mechanism of electrochemical oxidation of quercetin on vitreous carbon electrode using cyclic, differential and square wave voltammetry methods at different pH values.[4] As it follows from their study, quercetin strongly adsorbs on the electrode surface, and its surface is blocked by the final product of oxidation. The oxidation of catalytic 3, 4 – catechol group occurs first, at very low positive potentials, and is a semi reversible reaction, involving two protons and two electrons. Then, the oxidized hydroxyl group undergoes irreversible oxidation and can create intermolecular hydrogen bonds with an adjacent oxygen. The other two hydroxyl groups demonstrate a tendency to donate electrons and their oxidation is reversible.

A. K. Timbola et al. studied the electrochemical water-alcohol solutions of quercetin within the pH range of 2.2 to 9.2.[5] The study was carried out by means of cyclic voltammetry and UV-Vis spectroscopy. The cyclic voltammograms obtained for quercetin showed three oxidation peaks and one reduction peak depending on the conditions of the experiment. The first oxidation peak (at 0.15 V vs SCE at neutral pH) corresponds to the oxidation of catechol (ring B: 3′,4′-dihydroxyphenyl) to the corresponding orthoquinone (and its proton tautomers). The reduction peak corresponds to its reduction back to the catechol (E_red = 75 mV, reversible catechol/orthoquinone redox couple). Orthoquinone, a highly electrophilic molecule, was shown to be kinetically unstable, reacting rapidly with the nucleophiles of the medium (water and ethanol) as evidenced by the dependence of the cyclic voltammogram on the applied scanning speed. The two other oxidation peaks correspond to the oxidation of products formed by the reaction of the orthoquinone (and of its proton tautomers) with water and ethanol as shown by controlled-potential electrolysis and UV spectroscopy,[6] I.E. Mulazimoglu and E. Ozkan investigated the

![Chemical Structure of Quercetin](image)

**Figure 1.** The chemical structure of quercetin (quercetin: 3,3′,4′,5,7 – penta hydroxyflavone).

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electrochemical performance of quercetin on a vitreous carbon electrode modified with procaine and aminophenyl.\textsuperscript{[6]} They observed that at the first stage quercetin is oxidized electrochemically, whereas at the second stage it forms strong electrochemical bonded with the electrode surface. M. Medvedović-Kosanović et al. investigated the electrochemical properties of three structurally different flavonoids: catechin, quercetin, and rutin on a glassy carbon electrode surface at different pH values.\textsuperscript{[7]} It was found that in all the tested compounds oxidation of the 3, 4-dihydroxyl groups occurs in the B-ring (the first oxidation peak). The process is reversible, pH-dependent and includes transfer of 2 electrons and 2 protons. Electrochemical oxidation products of all the tested flavonoids strongly adsorbed on the surface of the electrode. H. R. Zare et al. oxidized quercetin electrochemically in 0.1 M phosphate buffer using the cyclic voltammetry method.\textsuperscript{[8]} They determined experimentally that the formal potential of quercetin relative to the saturated calomel electrode (SCE) amounts to 558 mV. The theoretical formal potential for quercetin in relation to SCE is 568 mV, which shows a high consistency of the experiment with the theory. D. Zieleńska and B. Pierożyński oxidized quercetin electrochemically using the cyclic voltammetry method on a vitreous carbon electrode, where the basic electrolyte was 0.1 M acetate buffer in 90% methanol.\textsuperscript{[9]} Impedance spectroscopy was also applied in the research. The study confirmed the cascade mechanism of quercetin oxidation, in which the initial stage was the oxidation of the hydroxyl groups of catechol, the intermediate product of the reaction, strongly adsorbed on the electrode. The paper presented also an original method of regeneration of vitreous carbon electrodes blocked by quercetin oxidation products. D. Zieleńska et al. studied also the process of adsorption and electrooxidation of two quercetin glucosides: quercetin 3-O-b-glucoside (Q 3-glc) and quercetin 4-O-b-glucoside (Q 40-glc).\textsuperscript{[10]} The methods applied in the research included cyclic voltammetry, impedance and UV-VIS spectroscopy, as well as HPLC-MS chromatography. The experiments were carried out in 0.1 M sodium acetate buffer and acetic acid in 90% methanol solution of methanol on a glassy carbon electrode surface. The results of the research provided new information on the mechanism of electrooxidation of two structurally different glucosides (quercetin derivatives). It was established that there is a close and important correlation between the structure of the flavonoid molecules and its antioxidant properties.

As previously reported, the constant magnetic field can affect electrochemical processes.\textsuperscript{[11–17]} The studies indicated that such changes are caused by the magnetohydrodynamic effect (MHD). The MHD effect is based on the impact of the Lorentz force, which induces the movement of the electrolyte and increases, or decreases, the transport of electroactive particles to the electrode. Magnetic fields acting on both the electrons and ionized atoms produce dynamic effects, one of which is volume movement of the liquid. The movement of the masses causes in turn modification of the fields. So we have to deal with the complex coupled system of the matter and fields. O. Vittori et al. studied the oxidation reactions of hexacyanoferate(II) and hydroquinone in KCl media on a platinum disc electrode using chronoamperometry under strong magnetic field (1,74 T). They concluded a drastic influence of the electrolyte dielectric constant on the limiting current.\textsuperscript{[18]}

Constant magnetic field may affect the efficiency and time of organic syntheses as well as the physicochemical parameters of materials.\textsuperscript{[19–20]}

2. Results and Discussion

Effects of constant magnetic field on chemical processes are a branch of science that is still developing. In the recent years, there have been many studies and numerous theories concerning magnetoelectrochemistry. Analysis of the published studies investigating the influence of constant magnetic field on chemical reactions allowed to find several types of magnetic field effects.\textsuperscript{[13,14,19–20]} Firstly, electrodeposition, electopolymerization, and some organic reactions occur faster under constant magnetic field. The surface of metal and alloy coatings obtained in the presence of magnetic field is smoother, and their grains are finer. Uniform magnetic field, applied during electrodeposition, improves corrosion resistance of some coatings. The effects of constant magnetic field have not been fully investigated yet. There is no uniform model for controlling constant magnetic field in electrodeposition. Nevertheless, the studies described in this paper demonstrate that constant magnetic field can be widely used in electrochemistry and electrodeposition industries. This is the reason why it is useful to study those phenomena and work on further development of magnetoelectrochemistry. The quercetin oxidation process occurred in three stages, as evidenced by the three anodic oxidation peaks recorded on the CV curves. The oxidation of the catechol OH groups in the B-ring was the easiest (Figure 1) (within the lowest range of potentials where the antioxidant activity is the highest). Then, the OH group in the C-ring was oxidized. The oxidation process of the OH groups in the A-ring took place at the highest potentials. As it can be seen on the CV curve (Figure 2), constant magnetic field increased the efficiency especially of the third stage of the quercetin electro-
oxidation process, i.e. the stage at which oxidation of the OH groups in the A-ring was the most difficult.

The value of the current density is 0.545 mA/cm² ($B = 0$ T) and 1.227 mA/cm² ($B = 0.6$ T). Under the influence of a constant magnetic field, the current density value of the first peak of quercetin C-ring oxidation (at the lowest potentials) increases slightly while the potential of the peak is shifted towards the negative values. This is due to the catalytic effect of a constant magnetic field. The potentials of the second and third oxidation peaks of C and A rings, in the constant magnetic field, are shifted towards positive values. At the same time, the anode current density of the third oxidation peak increases (ring A). The reason for these changes may be the fact that due to the catalytic effect of a constant magnetic field, the increasing amount of oxidized quercetin molecules may cause increased adsorption on the surface of the gold electrode (a change of its surface), and thus also increase the overpotential of process activation. Under the influence of the magnetic field, unstable intermediates of the compound may also be formed, which are a probable reason for shifting the potential of the second and third quercetin oxidation peaks towards positive values. The simple dependence of anode peak currents on the square root of the potential rise rate was linear and passed through the origin of the coordinate system. The above evidences that the oxidation processes of antioxidants are the processes controlled by diffusion. For various values of magnetic induction $B = 0.2$ T; 0.4 T; 0.6 T; 0.8 T; the dependence of the peak anode current density ($I_0$) on magnetic induction $B$ for the third stage of the quercetin electrooxidation process was obtained (Figure 3).

Up to the value of $B = 0.6$ T, constant magnetic field caused an increase in the efficiency of quercetin oxidation, which then began to decrease as a result of the magnetohydrodynamic forces, or too large values of the Lorentz force. The highest value of the oxidation peak currents was obtained at the magnetic induction value $B = 0.6$ T for the third stage of quercetin oxidation process.

The beneficial effect of the magnetic field on the efficiency of quercetin oxidation was confirmed by the results of impedance spectroscopy measurements. The impedance characteristics were recorded at the potential value corresponding to the third stage of the quercetin electrooxidation process (peak 3 in Figure 4) both without and with the presence of CMF. In both cases, a single and distorted semicircle is observed in the Nyquist impedance plots within the whole frequency range (see Figure 4a). The obtained characteristics were fitted by the electrical equivalent circuit presented in Figure 4b. The circuit exhibits a parallel combination of faradaic charge-transfer resistance $R_{ct}$ and double layer capacitance (represented here as constant phase element CPE for distributed capacitance) joined in series with uncompensated solution resistance $R_s$. The mean values of each element together with the standard deviation are summarized in Table 1.

Based on the obtained results it can be stated that CMF changes the diameter of the semicircle in the Nyquist plot, that represents the charge transfer resistance, thus CMF affects the kinetics of quercetin electrooxidation as well. In the presence of CMF, the charge transfer resistance parameter ($R_{ct}$ in Table 1) reaches a value twice lower as compared to that obtained without CMF. Such results indicate the positive effect of CMF on quercetin electrooxidation. Thereby, EIS results confirm the observation made on the basis of the voltammetric results described above.

The electrolysis of quercetin with controlled potential was carried out. The process was carried out in 200 seconds without the use of a constant magnetic field and in the magnetic field with induction values of 0.2 T, 0.4 T, 0.6 T, 0.8 T and 1 Tesla at the potentials of the third peak of the quercetin ring A

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**Table 1. Resistance and capacitance parameters for the third stage of the quercetin electrooxidation process (at peak NR) on gold electrode without and with the presence of CMF, obtained from the fitting procedure.**

| Condition      | $R_s/\Omega$ cm² | $CPE/\Omega$ cm² | $n$ | $R_{ct}/\Omega$ cm² |
|----------------|------------------|------------------|-----|---------------------|
| without CMF    | 35.0 ± 1.2       | 154.6 ± 8.1      | 0.850 ± 0.022 | 740.5 ± 9.9        |
| with CMF       | 38.6 ± 1.5       | 146.2 ± 7.7      | 0.876 ± 0.029 | 369.0 ± 8.3        |

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Figure 3. The dependence of the peak anode current density $I_0$ (surface area) on magnetic induction $B$ for the third stage of the quercetin electrooxidation process in the CV method.

Figure 4. a) Impedance characteristics presented as Nyquist and Bode plots for the third stage of the quercetin electrooxidation (at peak NR) on the gold electrode without and with the presence of CMF (the dots correspond to experimental data, the solid lines correspond to representation of the data according to the equivalent circuit); b) equivalent circuit for the quercetin electrooxidation at peak NR.
oxidation, for the individual fields. HPLC spectra were prepared for the solutions. The use of HPLC confirmed the results of tests performed using the CV method. The HPLC spectra obtained for the solutions after electrooxidation of quercetin at different magnetic induction values are shown in Figure 5.

On the basis of the obtained chromatograms, the areas under the peaks demonstrating the concentration of quercetin remaining after the reaction were calculated. The minimum value of the area under the peaks was observed at $B = 0.6$ T, and therefore the yield of the reaction was the best at that magnetic induction value. This is confirmed by the result obtained by CV. It can be observed on the graph, presenting the dependence of the area of the peaks in the chromatogram on the value of magnetic induction, at which the electro-oxidation process was conducted (Figure 6).

At the beginning of the paper, the antioxidant properties of quercetin were referred to. The antioxidant capacity of quercetin was investigated by means of spectrophotometry using the DPPH (1.1-diphenyl-2-picrylhydrazyl) free radical (Figure 7).

It was also checked how constant magnetic field affects the enhancement of these properties. The spectrophotometric measurements were conducted at a wavelength of $\lambda = 517$ nm. In the reaction with quercetin, the radical created the reduced form. The decrease in absorbance was proportional to the amount of the oxidized form which remained in the solution. The method used in the research is commonly used and known as a percentage reduction of DPPH.[21–22] It is sometimes referred to as “inhibition” or “quenching”, and defined by the following equation (1):

$$ Q = \frac{1}{2} \left( \frac{A_0 - A}{A_0} \right) \times 100\% $$

where: $A_0$ – absorbance of the DPPH radical solution, $A$ – mean value of absorbance of the tested antioxidant-containing solution.

The Q value denotes the ability of the tested antioxidant to counteract oxidation reactions. For the reactions taking place without exposure to constant magnetic field, this ability to counteract oxidation amounted to 90.2 %, while under exposure to constant magnetic field it reached 97.8 %. Constant magnetic field caused a 7.8 % increase in the ability of the tested antioxidant to counteract oxidation.

The physical essence of the effect of a constant magnetic field on various research objects is exerting mechanical forces on the moving charged particles (electrons or ions) that are a specific form of convection current or conduction. In an electrolyte solution, constant magnetic field affects both electrons and ionized atoms inducing the dynamic effects. One of them is volume movement of the liquid. The movement of the masses causes in turn modification of the fields. Thus, we deal with the complex coupled system of the matter and fields. The most important impact of the CMF on the electrochemical processes is the magnetohydrodynamic effect (MHD). The driving force behind this effect is the generated Lorentz force $F$ (2):

$$ F = \frac{\mu_0 N I L}{2\pi} $$
\[ F = j \times B \]  

where: \( j \) – limiting current density [A·m\(^{-2}\)], \( B \) – magnetic induction [T].

For configuration \( B \parallel P_e \) i.e. \( B \parallel j \) (where: \( P_e \) – area of the electrode [m\(^2\)], \( j \) – limiting current density [A·m\(^{-2}\)]) this is a macroscopic effect. It induces additional convection, causing an increase in mass transport towards the surface of the electrode. This causes the occurrence of laminar electrolyte flow towards the surface of the working electrode, which reduces the thickness of the diffusion layer and causes an increase in the gradient of potential-forming ions concentration. Generally speaking, the solution mass flow from and to the electrode may occur by migration, diffusion and convection. Diffusion is an important phenomenon occurring always in the process of electrolysis. The ion mass transfer can be extended with convection and the phenomenon can be defined as the convection diffusion. Between the surface of the electrode and the electrolyte mass there is a layer of the solution of convection and the phenomenon can be defined as the electrolysis. The ion mass transfer can be extended with important phenomenon occurring always in the process of diffusion. Diffusion is an important phenomenon occurring always in the process of diffusion. Diffusion is an important phenomenon occurring always in the process of diffusion. Diffusion is an important phenomenon occurring always in the process of diffusion. Diffusion is an important phenomenon occurring always in the process of diffusion.

The increase in magnetic induction \( B \) correlates also with increased thickness of the Navier-Stokes hydrodynamic layer \( \delta_H \), which determined the changes in the electrolyte flow under exposure to CMF.

For comparison, the paper presents a study of the effect of constant magnetic field (CMF) on the basic processes of Co–Mo, Co–W, Co–Mo–W alloys and Co electrodeposition.\(^{[13]}\)

On the basis of the coulometry (C) measurements, the relationship between the charge \( Q \) flowing in the electrolyte and magnetic induction \( B \) were determined for Co, specific Co–Mo, Co–W and Co–Mo–W alloys. Figure 8 indicates that the more complicated the structure of the metallic coating, i.e. Co, Co–Mo, Co–W, Co–Mo–W, the higher the value of magnetic induction \( B \) required to achieve maximum effect of the CMF on the charge \( Q \).

At present it seems that all the various effects of magnetic fields in electrochemistry are somehow related to mass transport. In the process of Co–Mo, Co–W, Co–Mo–W alloys and Co electrodeposition, the Lorentz force was generated as a result of the exposure to CMF. This force induced MHD effects in the solution, thus causing electrolyte movements, with the resultant depletion of the Nernst diffusion layer and appearance of a new Navier-Stokes hydrodynamic layer, which determined the velocity of electroactive molecules flow to the working electrode.

We also investigated the effects of magnetic field on the Kabachnik-Fields reaction.\(^{[18]}\) The reaction, which is shown in Figure 9, was carried out in the refluxing acetonitrile in the absence and in the presence of 1-T magnetic field, and the reactions were monitored by 31P NMR spectra. There were two types of magnetic field effects on Kabachnik-Fields reaction: improved yields of all the isolated aminophosphonates (3Aa–3Bf) and increased conversion rates as shown in Figure 10. The reason for the accelerating effect of magnetic field may be due to the orientation of substrate molecules. Magnetic field forced
the direction of molecules and reduced the distance between them.

3. Conclusions

The aim of the study was to examine the effect of constant magnetic field (CMF) on the course of electrochemical reactions of quercetin, a compound belonging to the flavonoid group. Quercetin belongs to a group of flavonoids, which are present in fruit, vegetables and wine. The magnetic induction $B$ with values from zero to 0.8 Tesla, applied in the studies, was directed in parallel to the surface of the working electrode, i.e. perpendicular to the direction of the current flow $j$. The quercetin oxidation process occurred in three stages, as evidenced by the three anodic oxidation peaks recorded on the CV curves. Increasing the efficiency of the third stage of quercetin oxidation process enables to conclude that the whole process can be considered to be catalyzed by the constant magnetic field. The HPLC method confirmed the results of tests performed using the CV method. The antioxidant capacity of quercetin was investigated by means of spectrophotometry using the DPPH (1,1-diphenyl-2-picrylhydrazyl) free radical. It was also checked how constant magnetic field affects the enhancement of these properties. For the reactions taking place without exposure to constant magnetic field, this ability to counteract oxidation amounted to 90.2%, while under exposure to constant magnetic field it reached 97.8%. Constant magnetic field caused a 7.8% increase in the ability of the tested antioxidant to counteract oxidation. As observed previously the constant magnetic field can affect electrochemical processes. The studies indicated that such changes are caused by the magnetohydrodynamic effect (MHD). The MHD effect is based on the impact of the Lorentz force, which induces the movement of the electrolyte and increases or decreases the transport of electroactive particles to the electrode. The Nernst diffusion layer ($b_\text{D}$) was reduced, whereas the new Navier-Stokes hydrodynamic layer ($b_\text{N}$) appeared.

Experimental Section

The aim of the study was to examine the effect of constant magnetic field (CMF) on the course of electrochemical reactions of quercetin, a compound belonging to the flavonoid group. The voltammetric curves were obtained by means of the CV method. A sample CV curve obtained during electrooxidation of quercetin without exposure to constant magnetic field ($B = 0$) and under exposure to a constant magnetic field of magnetic induction $B = 0.6$ T has been presented.

Cyclic voltammetry (CV) was the method applied in the studies of electrochemical reactions of quercetin. An Atlas 0531 Electrochemical Unit potentiostat was used in this method. Another element synchronized with the potentiostat was a three-electrode electrochemical cell made according to our own design containing (Figure 11): the working electrode (a gold disc) with an area of 0.1 cm$^2$, the auxiliary electrode (platinum, reticular) and the reference electrode (saturated, calomel). The working electrode was successively prepared mechanically (with the use of sandpapers, down to 4000 grade), chemically (using a chromic acid cleaning mixture) and electrochemically (polarization in a stock solution of 0.1 M NaClO$_4$ within the potential range of $-1$ V to $+2$ V, with a sweep rate of 100 mV/s$^{-1}$).

The magnetic induction $B$ with values from zero to 0.8 Tesla, applied in the studies, was directed in parallel to the surface of the working electrode, i.e. perpendicular to the direction of the current flow $j$. Under these conditions, the Lorentz force, arising as a result of the CMF effect, had the maximum value. The test solution of quercetin ($C_{15}H_{10}O_7$) was 302.24 g/mol, Sigma-Aldrich) (quercetin: 3,3',4,5,7-pentahydroxyflavone) used in the research had the concentration of 0.01 M. The primary electrolyte was sodium chloride (VII) (NaClO$_4$, $M_w = 140.46$ g/mol, POCH) at a concentration of 0.1 M in ethanol: water (1:1) solution. Electrochemical impedance spectroscopic (EIS) measurements were performed using a PGSTAT 128 potentiostat / galvanostat with FRA2 module (EcoChemie Autolab) operated with FRA software (v. 4.9). Electrochemical impedance characteristics were registered at peak NR potential by applying a sinusoidal signal of ±5 mV within the frequency range from 0.06 Hz to 10 kHz. The EIS measurements were carried out without and with the presence of CMF in triplicate. Chromatographic (HPLC) studies were also conducted. The mobile phase was a mixture of acetonitrile and sodium dihydrogen phosphate (V) (45:55 v/v). The flow rate was 1.2 ml/min, the wavelength 202 nm. Chromatographic separation was conducted using an Alltima C18, 250 mm × 4.6 mm column, Grace. Spectrophotometric studies were carried out with the use of a VIS Metertech SP-830Plus spectrophotometer.

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Conflict of Interest

The authors declare no conflict of interest.

Keywords: quercetin · constant magnetic field · Lorentz force
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