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Enhanced electrochemical activity of regenerated expanded graphite electrode after exhaustion in the process of phenol oxidation

J.M. Skowroński*, P. Krawczyk

Poznań University of Technology, Institute of Chemistry and Technical Electrochemistry, ul. Piotrowo 3, 60-965 Poznań, Poland

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ABSTRACT

The presented paper deals with the cyclic electrochemical oxidation of phenol occurring in alkaline solution on the electrode made of expanded graphite (EG). Passive oligomer layer produced on the electrode surface during the first cycle of phenol oxidation brings about a dramatic decrease in electrochemical activity of the EG anode in the subsequent cycles. It was shown that electrode spent during three cycles of phenol oxidation could be thermally regenerated at 400 °C to such an extent that its original electroactivity is significantly exceeded. The time of heat treatment exerts a strong influence on the properties of regenerated graphite. The increase in electrochemical activity of expanded graphite electrode after the regeneration process is attributed to enhanced activity of carbon–carbon composite formed due to carbonization of oligomer film covering the surface of exhausted graphite. The physicochemical features of the oligomer/EG composite were studied using electrochemical technique, scanning electron microscopy, thermal analysis, X-ray photoelectron spectroscopy (XPS) and BET measurements.

1. Introduction

Electrochemical oxidation of phenol seems to be effective and attractive method from the economical point of view of removal of phenolic compounds from waste water [1,2]. The decrease in the reaction efficiency of phenol oxidation with time is a key disadvantage of all electrochemical methods reported in the literature [1–14]. For this reason, after some time of reaction the exhausted material (adsorbents, electrodes) must be replaced for new one. It is obvious that in the case of periodic process the restoration of the original activity for spent material is cheaper than the use of a fresh one. Because of this, much effort is made to find out new materials and methods enabling multiple use of material in phenol oxidation. The main drawback of electrooxidation of phenol is an inactivation of electrode material by passive layer composed of polymer products of phenol oxidation. The problem of the electrode inactivation has been reported for many types of electrodes, e.g., platinum [3–5], platinum/gold [6], carbon fiber [7], glassy carbon [8] and nickel modified glassy carbon electrode [9]. Using the method of cyclic voltammetry the authors of papers [3–9] have observed a dramatic decrease in electrochemical activity after the first oxidation run. This phenomenon has also been observed by us for exfoliated graphite (EG) based electrode [12–14]. From the above the conclusion can be drawn that there are two parallel targets in electrooxidation of phenol: searching for a new anode material representing both a high oxidative activity and resistance against inactivation for a long time as well as attempting to find an easy method for regeneration of exhausted electrode material allowing the reduction of the process costs.

When graphite intercalation compound (GIC), in which intercalate is inserted between the carbon layers of graphite, is exposed to heat, an abrupt evaporation or decomposition of intercalate occurs simultaneously even in a hundred times expansion of graphite along the *c*-axis. The material generated in this way is named expanded graphite (EG) [15–17]. As compared to the host graphite, EG is characterized by much more developed surface area, higher adsorption capability, a higher resistance against the heat and flame and a low density. These properties of EG made possible its application as sorbents of heavy oils and other heavy hydrocarbons from waste water [18,19], high temperature gaskets, seals, packings, fire extinguisher agents, thermal insulators and composite filler [15].

Most of papers reporting carbon materials deal with removal of phenol with active carbons (ACs), which, in general, indicate a high adsorptive capability [20–27]. The release of phenol from porous structure of ACs is preferably performed using thermal methods [20–23,25,27]. During the heat treatment of carbon material, on increasing temperature a removal of adsorbed organic compounds proceeds effectively according to the following steps: evaporation of water, thermal desorption of volatile compounds, pyrolysis and the formation of carbon char (carbonization).

In the present work we report on the thermal enhancement of electrochemical activity of expanded graphite, previously coated

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* Corresponding author. Tel.: +48 61 665 3641; fax: +48 61 665 2571.
E-mail address: jan.skowronski@put.poznan.pl (J.M. Skowroński).

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by oligomer products of phenol electrooxidation. The removal of phenol from alkaline solution is realized on electrochemical way, whereas thermal treatment is applied to regenerate EG coated by oligomer product of phenol oxidation. Thermal conversion of such an oligomer film into carbon/carbon composite (C/C composite) electrode, of activity significantly exceeding that of the original EG electrode, is shown for the first time. The criterion of electrochemical activity of EG electrode is the anodic charge associated with phenol oxidation. In such a way the usefulness of electrode for removal of phenol from waste water can be preliminarily estimated [6,8]. Electrooxidation of phenol is carried out in alkaline solution using cyclic voltammetry technique. Electrochemical measurements were complemented by SEM, XPS, TG and DTA analyses.

2. Experimental

2.1. Preparation of expanded graphite

Expanded graphite used as the electrode material was prepared by thermal decomposition of graphite intercalation compound with sulphuric acid (H_2SO_4–GIC) at 800 °C for 5 min. A scanning electron micrographs of expanded graphite and thermally modified exhausted EG are presented in Fig. 1. H_2SO_4–GIC was prepared by anodic oxidation of graphite in 18 M H_2SO_4, according to the procedure described earlier [28]. The synthesized H_2SO_4–GIC was rinsed with water and subjected to decomposition. The specific surface area and average pore diameter of expanded graphite (before and after electrochemical and thermal treatments) were determined from the isotherms obtained by N_2 adsorption at 77 K. The attained data are given in Table 1.

2.2. Preparation of electrode and electrochemical measurements

The cyclic voltammetry measurements with a scan rate $0.1\,\mathrm{mV\,s^{-1}}$ were performed in the potential range from the rest potential of electrode (E_R) to 0.8 V using 0.5 M aqueous solution of KOH containing 0.1 M phenol. Similar to experiments reported in [3–6], a relatively high concentration of phenol was used to avoid the influence of the change in phenol concentration at the electrode/electrolyte interface on the kinetics of electrochemical process upon cycling. Because preliminary experiments showed that for a chosen concentration of phenol stirring the solution had no effect on the character of voltammograms recorded during three cycles, further measurements were performed in stationary electrolyte. After starting the measurements, the potential was swept from E_R in the positive direction. All the potentials on the presented voltammograms are referred to the Hg/HgO/0.5 M KOH electrode. In each case the mass of working electrode was equal to 30 mg.

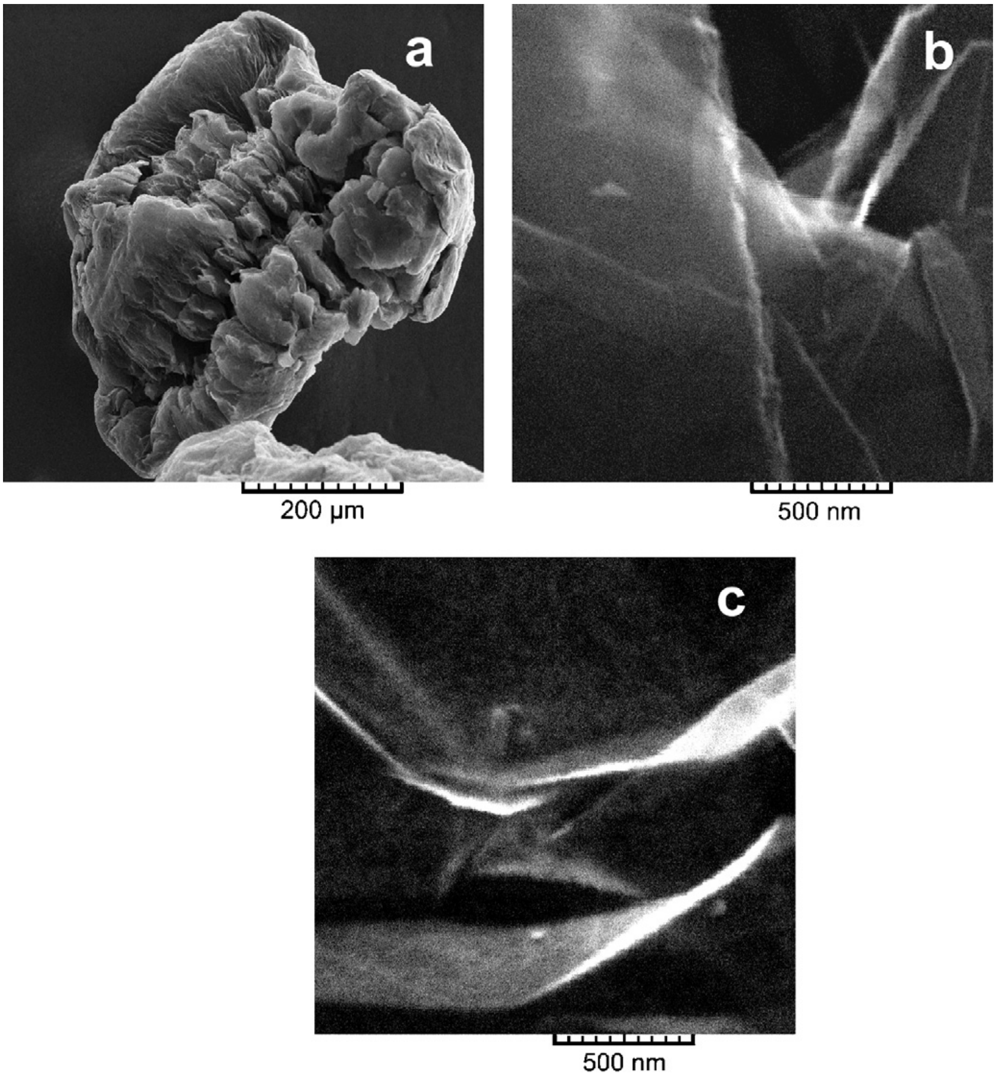

Fig. 1. SEM micrographs recorded for original EG (a and b), exhausted EG after regeneration at 400 °C for 3 h (c).

Table 1
Specific surface area and average pore diameter of investigated samples.

	Original EG	EG after 1 cycle of phenol oxidation	Exhausted EG after regeneration at 400 °C for 2 h	Exhausted EG after regeneration at 400 °C for 3 h
Surface area ($m^2 g^{-1}$)	30.9	7.5	21.8	20.4
Average pore diameter (nm)	9.0	9.2	10.4	11.7

The powder type working electrodes were prepared according to the following procedure. Expanded graphite was placed in a pocket made of porous polymer material, in which graphite rod (5 mm in diameter), playing a role of current collector, was beforehand inserted. A similar graphite rod was used as a counter electrode. The reference Hg/HgO/0.5 M KOH electrode was connected to the solution under investigation by a Luggin capillary. Before starting measurements the working electrode was equilibrated at an opened circuit for 1 h. After the electrochemical process of phenol oxidation, EG sample was taken out from the electrolytic cell and rinsed with distilled water and air-dried. Electrochemical measurements were performed using AUTOLAB potentiostat–galvanostat (model PGSTAT 30). Phenol used as reagent was purchased from LACHEMA (puriss. >99.5 wt.%).

2.3. Thermal regeneration

Thermal regeneration of exhausted EG was carried out in muffle furnace at temperature 400 °C in air. The process of regeneration started after EG was inserted into cold furnace and sample was kept at a chosen temperature for 2 and 3 h.

2.4. XPS and TG analysis

The changes in the chemical composition of the regenerated graphite surface were determined with an X-ray photoelectron spectroscopy (XPS) (ESCALAB 210 spectrometer; VG Scientific, East Grinstead, UK) using a non-monochromatized Al Kα radiation (1486.6 eV). The attained XPS spectra were analyzed by performing Shirley-type background subtraction and using Gaussian–Lorentzian mix function of 70:30% ratio. The binding energy scale was corrected by referring to the graphitic peak at 284.5 eV.

Thermogravimetric analysis (TG) and differential thermal analysis (DTA) were done in air at a rate of 5 °C min^{-1} using a Setaram TGA 92 apparatus.

3. Results and discussion

3.1. Electrochemical oxidation of phenol on original EG

Fig. 2 presents cyclic voltammograms recorded in 0.1 M solution of phenol in 0.5 M KOH for the original EG. A large anodic peak corresponding to phenol oxidation is observed at the potential around 0.43 V for the first cycle. As seen from this figure, anodic charge decreases steeply after the first oxidation cycle. This feature has been related to the loss of electrochemical activity due to the formation of thin layer of passive oligomer film on the EG surface during the first oxidation cycle [10] (anodic charges measured during electrochemical phenol oxidation are given in Fig. 3). Such an explanation is consistent with the fact that no reduction peak is recorded after the polarization direction is reversed. It is known that one of the possible pathway of phenol electrooxidation can proceeds according to the formation of passive polymer products, which makes electrode inactive [3–10]. The evidence for the formation of passive layer on the EG particles is given in Table 1, where a four-fold decrease in BET surface area is shown for EG after one cycle of phenol oxidation. An average pore diameter changed insignificantly. It suggests that the oligomer film almost tightly coating the EG surface is itself porous. However, the volume of pores of this film contributing to the BET surface is considerably lower than that of the original EG. The presence of non-conductive layer on EG surface, responsible for a decrease in BET surface area, accounts for the worsening of electrochemical activity in the second cycle. On the other hand, the increase in BET surface area after the regeneration of exhausted EG to values not attaining that for the original EG might be elucidated in terms of closing micro- and mesopores in graphite flakes by oligomer layer.

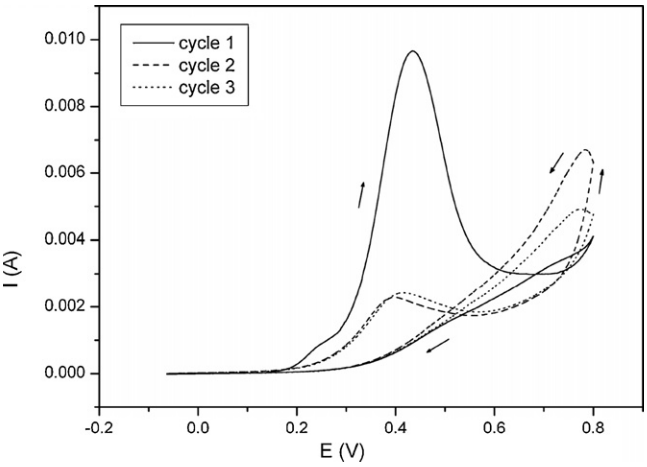

Fig. 2. Cyclic voltammograms recorded in 0.1 M phenol in 0.5 M KOH for original EG. Potential range: $E_R \leftrightarrow 0.8$ V, $E_R = -0.64$ mV, scan rate: 0.1 mV s^{-1}.

The process of phenol electrooxidation on EG electrodes starts from the transformation of phenol ions into phenoxy radicals. During the subsequent oxidation phenoxy radicals can be transformed into the quinonic compounds, such as hydroquinone and benzo-

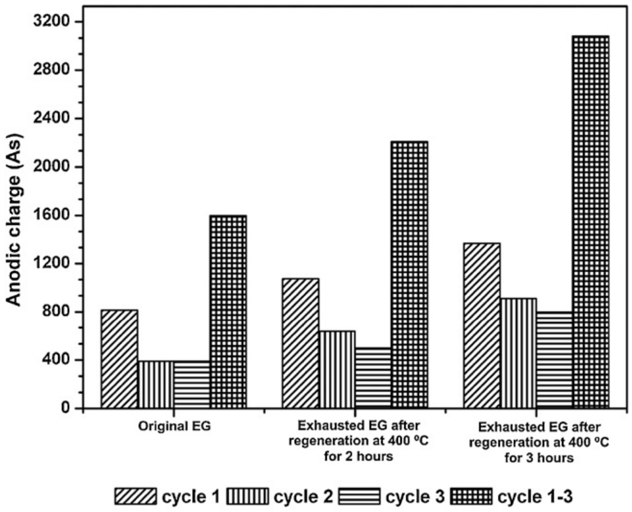

Fig. 3. Comparison of anodic charges recorded during electrochemical phenol oxidation for original and modified EG anodes. Original EG (1), Exhausted EG after regeneration at 400 °C for 2 h (2), Exhausted EG after regeneration at 400 °C for 3 h (3).

quinone, followed by the further oxidation to carboxylic acids and finally to CO_2 and water. As an alternative route, phenoxy radicals undergo coupling reactions which initiate the process of polymerization. The regarded process has a multi-step character, and results in the formation of a variety of compounds of different structures. Depending on the type and number of substitutes in aromatic ring of phenol molecule, among the possible products of the phenoxy radicals coupling reactions can considered polymer radicals, ether and quinine-like oligomeric compounds, polyphenol derivatives. The particular description of the mechanisms of oligomer formation during the phenol or/and its derivatives electrooxidation are reported in the literature [4,29].

A considerable decrease of anodic peak at 0.43 V during the forward scanning and the growth of anodic current at potentials higher than 0.55 V during the backward scanning, both noted in the second and third cycle (see Fig. 2), allow us to assume that electrochemical properties of EG are markedly altered after the first cycle of phenol oxidation. The current loop formed over 0.55 V may be attributed to the consecutive reactions of phenol oxidation as well as to the oxidation reaction of oligomer products formed on the EG surface during the first cycle of phenol oxidation. The latter presumption is corroborated by the fact that the charges recorded in the potential range of the loop are considerably higher for the second and third cycle as compared to that noted for the first cycle.

3.2. Electrochemical oxidation of phenol on regenerated EG

The presence of inactive oligomer film on the surface of graphite electrode, bringing about an abrupt decrease in electrochemical activity in subsequent steps of phenol oxidation, implied the need for the regeneration of exhausted electrode.

3.2.1. Electrochemical oxidation of phenol on EG regenerated at 400 °C for 2 h

Fig. 4 shows cyclic voltammograms recorded in the phenol-containing electrolyte for EG regenerated at 400 °C for 2 h. Before thermal regeneration the EG anode was subjected to three cycles in the process of phenol oxidation ($E_R \leftrightarrow 0.8$ V). From comparison of CV curves presented in Figs. 2 and 4 one can see that the kinetics of phenol oxidation changes significantly after the regeneration of exhausted electrode. Thermally treated graphite exhibits higher electrochemical activity than as compared to the original graphite. Anodic charge calculated for the first cycle of phenol oxidation on regenerated electrode is about 32% higher than that for the original EG (see Fig. 3). The enhancement of electrochemical activity after the process of regeneration is also observed for the subsequent cycles. The peak charge for the third cycle of regenerated sample significantly exceeds the charge calculated for the second cycle of the original EG. Moreover, the current loop in the potential range 0.55–0.8 V still observed for the third cycle of the original EG (see Fig. 2), disappears completely for regenerated EG (Fig. 4). This observation indicates that during heat treatment of EG/oligomer composite at 400 °C for 2 h the decomposition of oligomer film occurs with simultaneous chemical as well as physical modification of the EG surface. It is likely that such a modification results in the changes in both porosity and chemical composition due to simultaneous oxidation of exfoliated graphite substrate and the formation of carbon film onto the EG surface during carbonization of oligomer film. This assumption agrees with the results of thermal analysis. As can be seen from thermogravimetry (TG) and differential thermal analysis (DTA), expanded graphite is thermally resistant in air up to 650 °C where its burning-up starts to give a large exothermic peak at 780 °C (Fig. 5a). For expanded graphite coated by the product of phenol oxidation three exothermic peaks at 297, 384 and 460 °C are noted on DTA curve in the temperature range 20–500 °C, whereas burning-up starts at about 530 °C (Fig. 5b). These effects, coincident with three slopes of the weight loss on TG curve, are indicative of a three-step decomposition/carbonization process of EG/oligomer composite. The reaction of thermal decomposition/oxidation of EG coated with the products of anodic oxidation of phenol suggests that the oligomer coating is composed of multiple layer of different thermal stability (Fig. 5b). It seems plausible that the thin oligomer film produced at lower potentials directly onto the graphite surface (a small anodic peak noted at about 0.25 V on CV curve; Fig. 2) differs in the chemical and porous structure from the outer one formed at higher potentials onto the inner oligomer substrate. The outer layer protecting the inner one against oxidative action of air during the TG/DTA measurements is formed at higher potentials (a large anodic peak with the maximum about 0.45 V in Fig. 2). The larger peak can be attributed to a thicker layer. Based on such an assumption it is likely that a higher exothermic peak recorded at 297 °C on the DTA curve arises form the oxidation of the outer layer with a high porosity due to the effect of overoxidation occurring at potentials higher than 0.55 V, whereas a smaller exothermic peak observed at higher temperature (460 °C) corresponds to the oxidation of the inner oligomer film firmly attached to the EG surface. At this stage of investigation the origin of a trace exotherm at 384 °C is difficult for explanation, but the contribution of intermediate products of phenol oxidation encapsulated in the oligomer matrix is worthy of considering in the future. When the oxidation of the inner oligomer film occurs the EG surface is locally burnt resulting in the development of porosity, especially at the edge regions of the EG fakes. The EG surface with a high porosity may resemble that of activated carbons. In consequence, the susceptibility to burning increases. Accordingly, the main exothermic peak related to burning the EG/oligomer composite is shifted over 180 °C towards lower temperature as compared to that for the original EG. Based on the TG and DTA data it is reasonable to point out that carbon product deposited onto the EG surface undergoes subsequent oxidation upon treatment in air atmosphere. The regeneration temperature of 400 °C is justified by these results and is in agreement with data reported elsewhere for activated carbons covered with adsorbed phenolic compounds [20,23,24,26].

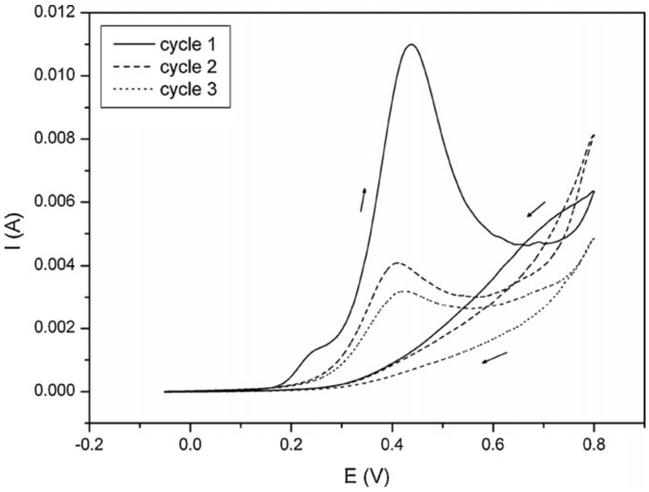

Fig. 4. Cyclic voltammograms recorded in 0.1 M phenol in 0.5 M KOH for exhausted EG after regeneration at 400 °C for 2 h. Potential range: $E_R \leftrightarrow 0.8$ V, $E_R = -0.51$ mV, scan rate 0.1 mV s^{-1}.

3.2.2. Electrochemical oxidation of phenol on EG regenerated at 400 °C for 3 h

To reveal the influence of the regeneration time on the enhancement of electrochemical activity of regenerated EG towards phenol oxidation, the operation time was expanded from 2 to 3 h. Fig. 6 shows cyclic voltammograms recorded in phenol containing elec-

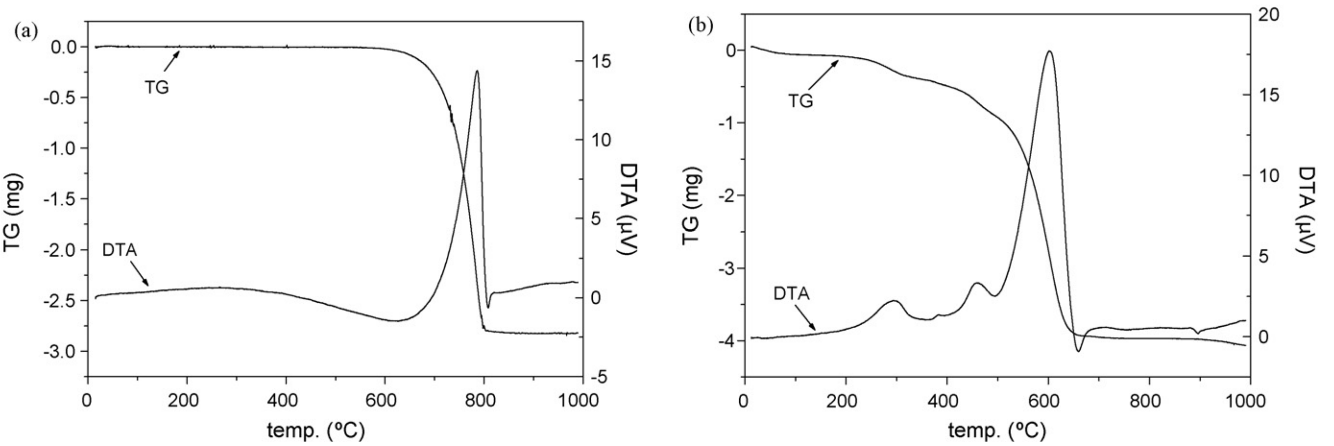

Fig. 5. TG and DTA curves recorded for original EG (a), original EG coated by the product of phenol oxidation during 3 cycles (b). Sample weight: (a) 3.2 mg, (b) 4.2 mg. Air atmosphere, heating rate: 5 °C min^{-1}.

trolyte for EG regenerated for 3 h. From comparison of Figs. 4 and 6 it is clear that the anodic charge associated with phenol oxidation increases considerably as the regeneration time is 1 h prolonged. For the first cycle of phenol oxidation, anodic charge of exhausted EG after heat treatment for 3 h becomes 68% higher than that for the original EG and is about 28% higher than that for the EG regenerated during 2 h (see Fig. 3). The improvement of electrochemical activity of graphite electrode regenerated for 3 h is also noted for the subsequent cycles. The anodic charge measured for the second cycle of EG regenerated for 3 h is significantly higher than that for the respective cycle of sample regenerated for 2 h, and also exceeds the charge for the first cycle of the original graphite. Moreover, electrochemical activity measured in the third cycle for EG regenerated for 3 h is nearly equal to that estimated for the first cycle of the original EG. This result proves that the EG electrode obtained after 3 h of regeneration is attractive material from the point of view of a long-term electrooxidation of phenol.

The increase in electrochemical activity of expanded graphite electrode attained for a longer time of regeneration may be attributed to enhanced activity of C/C composite formed due to the process of carbonization of oligomer film coating the surface of exhausted graphite. The above suggestion is supported by SEM micrographs recorded for EG before and after the regeneration for 3 h (Fig. 1b and c). Under a high magnification it was possible to observe that a very smooth surface of the original graphite (Fig. 1b) changed to a spotted-surface of the regenerated graphite. This effect may be related to porous carbon layer existing on the expanded graphite substrate. It is reasonable to assume that during the process of regeneration, starting at a room temperature, oligomer layer undergoes the first exothermic conversion, accompanied by weight loss, at about 300 °C. After furnace attains 400 °C further weight loss of carbonized product takes place due to its reaction with air. It seems likely that during a rapid rise of temperature in the first stage of the process, a thin layer of oligomer present on basal planes of flakes is torn followed by its coalescence into carbonized beads of different size. They cover a dusty-like surface of graphite flakes, whereas the flake edges grow thicker due to more oligomer there deposited. It is worthy of noting that after three cycles of phenol oxidation the weight increase of 58.2%, resulting from the produced oligomer film on the EG electrode, was estimated. During the subsequent heat treatment of EG/oligomer composite in air at 400 °C, its weight decreased due to carbonization of oligomer and after 3 h of regeneration the weight loss was equal to 31.9%. In consequence, the resulting weight of the product was 7.7% higher as compared to that of the starting sample of exfoliated graphite, and this number can be related to carbon layer coating EG substrate. The existence of passive layer onto exhausted EG, which undergoes transformation to carbon coating, is confirmed by TG curves displayed in Fig. 5. In contrast to negligible weight loss of the original EG (Fig. 5a), the weight loss of EG coated by the product of phenol oxidation equals 11.9%, when measured in dynamic conditions in the temperature range 20–400 °C (Fig. 5b). According to the afore-proposed mechanism of regeneration, during carbonization of EG/oligomer, the EG surface is coated by porous carbon film with many cracks through which the EG surface is disclosed. Due to carbonization of the EG/oligomer, the EG surface is coated by porous carbon film of developed surface area and increased average pore diameter resulting from oxidative treatment increased. It might explain why surface area of C/C composite is distinctly higher than that of exhausted EG, but not as high as that of the original EG (Table 1). In the light of this, an enhanced activity of C/C composite produced owing to the regeneration process may be attributed to both the structural and chemical properties of carbon film and disclosed surface of graphite substrate. At this point it is reasonable to note that an average pore diameter, growing up significantly after regenerating exhausted EG, can make the transport of phenol molecules to active sites easier (see Table 1).

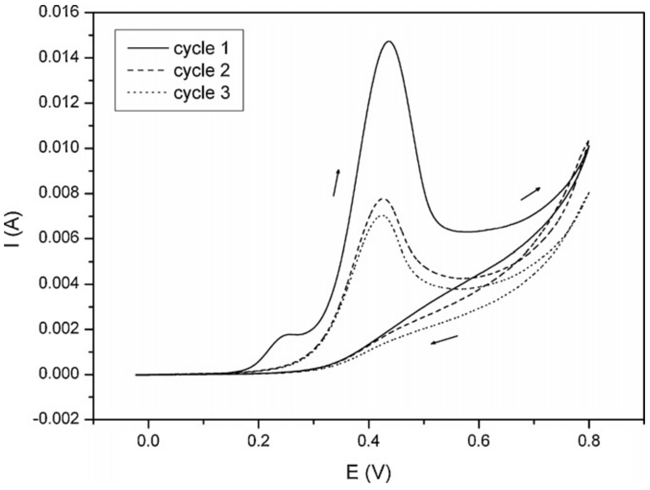

Fig. 6. Cyclic voltammograms recorded in 0.1 M phenol in 0.5 M KOH for exhausted EG after regeneration at 400 °C for 3 h. Potential range: $E_R \leftrightarrow 0.8$ V, $E_R = -0.25$ mV, scan rate 0.1 mV s^{-1}.

3.3. *XPS measurements*

In order to estimate the contribution of carbonized oligomer film to the improvement of electrochemical activity of regener-

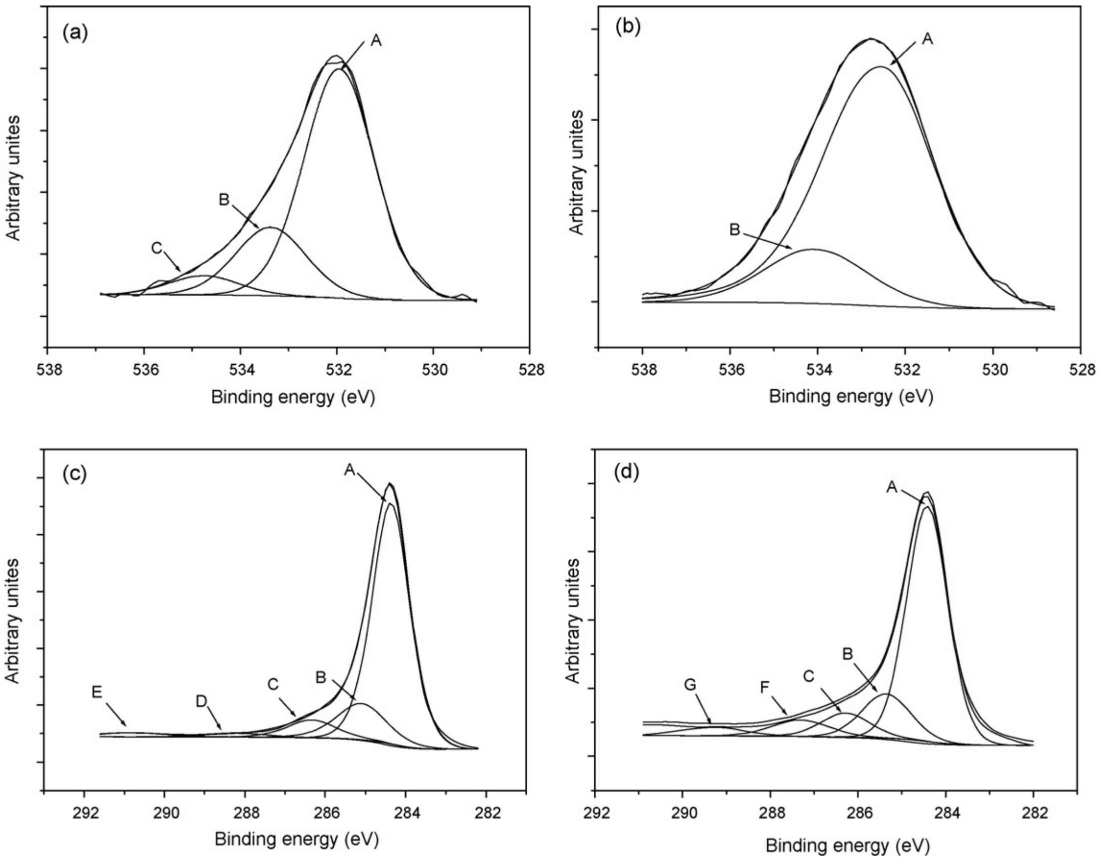

Fig. 7. XPS O 1s spectra for original EG (a), O 1s spectra for regenerated EG (b), C 1s spectra for original EG (c), C 1s spectra for regenerated EG (d).

ated EG, the XPS measurements for the original and regenerated EG sample were performed Fig. 7(a and b) presents deconvoluted spectra of O1s, whereas Fig. 7(c and d) displays deconvoluted spectra of C1s. The comparison of spectra C1s and O1s recorded for original EG and for regenerated EG/oligomer gives evidence of qualitative differences in the surface composition. The most pronounced difference from the point of view of enhancement of electrochemical activity is related to the surface oxygen concentration. Total oxygen content for regenerated sample (5.02 at.%) is over twice higher as compared to the original graphite (2.28 at.%). Based on the O1s/C1s atomic ratio calculated from the deconvoluted XPS spectra of both investigated samples, which can be applied to estimate the degree of oxidation, it can be pointed out that EG/C composite displays considerably higher degree of surface oxidation. The O1s/C1s atomic ratio for the EG/C sample was determined to be 0.055, whereas for the original EG was amounted 0.024.

The O1s spectrum for heat treated EG/oligomer shows that oxygen atoms are bounded with carbon in two different forms, i.e., C=O and C–O, which correspond to peak at binding energies of 532.7 eV (peak A) and 534.3 eV (peak B), respectively (Fig. 7b) [30,31]. It should be noted that the former bond is dominant. The O1s spectrum of the original EG (Fig. 7a) comprises three peaks at 532.0 eV (peak A), 533.4 eV (peak B) and 534.8 eV (peak C), which are probably associated with oxygen representing C=O, C–O species and adsorbed water, respectively [30–33]. The discrepancy in binding energies between the composite EG/C and pristine EG may suggests that O atoms attached to C atoms represent different oxygen containing functional groups.

Comparative analysis of C1s spectra recorded for the original EG and regenerated EG/oligomer provides additional information on chemical nature of the investigated surfaces. Generally, the signals in both deconvoluted C1s spectra could be divided onto two different parts. One of them represents C atoms pertaining to the graphite skeleton, i.e., graphitic peak at 284.5 eV (peak A) and the satellite at 290.9 eV (peak E), assigned to the π electrons delocalized within the aromatic structure of carbon [30–33]. The former effect is observed on both considered C1s spectra, whereas the latter signal appears only in the spectrum for the original EG. The second part of considered C1s spectra, with exception of binding energy peak at 283.5 and 282.8 eV (peaks B), corresponds to carbon atoms bounded with oxygen atoms representing surface functional groups. It is worth to note that in comparison with the original graphite, the C1s spectrum for regenerated EG/oligomer (Fig. 7d) consists of two new signals with binding energies of 287.5 eV (peak F) and 289.4 eV (peak G). These binding energy peaks can be identified as carbonyl and carboxylic or ester groups, respectively [30–32,34]. It cannot be excluded that the latter functionalities, especially carbonyl groups, are also present in the XPS spectrum for the original EG but at binding energy of 288.5 eV [34]. Another two pairs of peaks 286.3–286.4 eV (peaks C) and 285.0–285.5 eV (peaks B) of C–O species pertaining to the different functional groups, i.e., ether, alcohol, phenolic, metoxy, are common for both investigated samples [30–36].

Significantly lower concentration of graphitic carbon (55.36 at.%) for heat treated sample, as compared to the original EG (63.01 at.%), as well as the lack of signal arising from the π electrons in the aromatic structure associated with markedly higher contents of C ascribed to the functional groups (36.2 and 31.5 at.%), suggest that the regenerated EG is coated with the layer of non-graphitic carbon material. The above assumption leads to the conclusion that during heat treatment of EG/oligomer, simultaneously with the carbonization of oligomer, the formation process of oxygen functional group occurs, which probably accounts for the improvement of electrochemical activity in the process of phenol oxidation.

4. Conclusions

Because oligomer products formed onto the electrode surface during the process of phenol oxidation in alkaline solution resulted in the decrease in electrochemical activity of electrode, the regeneration of electrode material was performed in this work. Using the voltammetric method it was shown that graphite electrode spent during three cycles of phenol oxidation can be thermally regenerated to such an extent that its original electroactivity is significantly exceeded owing to the formation of a new material: carbon/carbon composite composed of EG substrate and the surface carbon film. The SEM analysis suggesting the formation of such a composite was supported by the results of XPS measurements. Compared to the original EG, the C1 s spectrum for heat treated EG/oligomer displayed lower content of the surface graphitic carbon with simultaneous increase of carbon atoms representing functional groups. This comparison allows the evidence of the existence of non-graphitic carbon layer on the surface of regenerated EG. On the other hand, an enrichment of the surface of regenerated composite material with surface oxygen group was proven by O1s spectra. Taking this into account, the surface oxygen functional groups may be recognized as a main factor responsible for the improvement of electrochemical activity of a new C/C composite material in the process of phenol electrooxidation. The time of heat treatment exerts a strong influence on the properties of regenerated graphite. The highest electrochemical activity was noted after the regeneration process carried out for 3 h. In this case, the regeneration efficiencies of 168% and 193% were reached for the first cycle and the total three cycles of phenol oxidation, respectively.

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