



Decolourisation of dyes under electro-Fenton process using Fe alginate gel beads

E. Rosales, O. Iglesias, M. Pazos, M.A. Sanromán*

Department of Chemical Engineering, University of Vigo, Isaac Newton Building, Campus As Lagoas, Marcosende 36310, Vigo, Spain

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ABSTRACT

This study focuses on the application of electro-Fenton technique by use of catalytic activity of Fe alginate gel beads for the remediation of wastewater contaminated with synthetic dyes. The Fe alginate gel beads were evaluated for decolourisation of two typical dyes, Lissamine Green B and Azure B under electro-Fenton process.

After characterization of Fe alginate gel beads, the pH effect on the process with Fe alginate beads and a comparative study of the electro-Fenton process with free Fe and Fe alginate bead was done. The results showed that the use of Fe alginate beads increases the efficiency of the process; moreover the developed particles show a physical integrity in a wide range of pH (2–8). Around 98–100% of dye decolourisation was obtained for both dyes by electro-Fenton process in successive batches. Therefore, the process was performed with Fe alginate beads in a bubble continuous reactor. High color removal (87–98%) was attained for both dyes operating at a residence time of 30 min, without operational problems and maintaining particle shapes throughout the oxidation process. Consequently, the stable performance of Fe alginate beads opens promising perspectives for fast and economical treatment of wastewater polluted by dyes or similar organic contaminants.

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1. Introduction

In recent years, more efficient and non-selective techniques like advanced oxidation processes (AOPs), offer effective and rapid alternative treatments for various contaminants [1]. The AOPs are based on the *in situ* generation of hydroxyl radicals ($\cdot\text{OH}$), a highly powerful oxidizing agent, and are effective in treatment of persistent organic pollutants in aqueous solutions until their overall mineralization [2]. Among the most promising AOPs for wastewater treatment, Fenton reaction with hydrogen peroxide and transitional metals, especially the ferrous ion, in an acidic aqueous system is particularly attractive and has been investigated in numerous studies [3].

Several studies demonstrated that one of the main drawbacks of Fenton treatment relies on the instability of H_2O_2 when it gets in touch with chemical species that are naturally in the environment [4,5]. To solve this problem, some research groups have reported that electro-Fenton oxidation offers significant advantages, without requirement for special equipment, and high efficiency in organic pollutants removal [6,7]. Electro-Fenton process is a promising technology that combines electrochemical reactions and Fenton process. In this hybrid process, the electrical current induces the reduction of ferric ions to form ferrous ions

and hydrogen peroxide which are needed for the Fenton reactions. Thus, they are generated *in situ* by the electrical current, avoiding their transport and continuous addition throughout the process due to its decomposition in the medium [2].

Therefore, in the electro-Fenton process the continuous electro-generation of H_2O_2 is achieved by O_2 reduction in the presence of dissolved Fe^{2+} . If there are Fe^{3+} species in the medium, they revert to Fe^{2+} by different reduction processes, involving H_2O_2 or organic intermediate radicals, as well as the direct reduction of Fe^{3+} on the cathode. This allows the propagation of the Fenton's reaction *via* a catalytic cycle [8].

It is well known that Fenton's reaction occurs at low pH values. Fe ions precipitate at pH higher than 4, reducing the degradation efficiency as well as difficulty post-treatment of Fe sludge after the reaction. In the literature, it is reported that the Fenton reaction has the highest efficiency when the pH is between 2 and 4 [9]. Zhou et al. [10] found that the highest electro-Fenton activity in the methyl red degradation was attained under pH 3, while at pH 5, the level was only 63.8% of that of dye removal at pH 3. Similar effect was detected in the degradation of other organic compounds. Li et al. [11,12] studied the degradation of 2,4-dichlorophenol in a wide pH range from 1 to 9 and determined that the maximal efficiency is obtained at pH 2.5–3. Masomboon et al. [13] studied the degradation of 2,6-dimethylaniline at several pHs (from 1 to 4) and they confirmed that pH has a sharp influence on the electro-Fenton reaction with a maximum reaction rate at pH 2. These results are in agreement with early reports [14,15].

* Corresponding author. Tel.: +34 986 812383; fax: +34 986 812380.
E-mail address: sanroman@uvigo.es (M.A. Sanromán).

Therefore, the solution pH provoked the change in the speciation of the Fenton's reagents reducing the oxidation reactions, thus an acidic pH is required in order to increase the efficiency of Fenton process. To overcome the narrow pH of the Fenton and electro-Fenton process that limits the wide application of this technology in the wastewater treatment, the utilization of heterogeneous Fenton reagents such as metal oxides [16], Fe@Fe₂O₃/ACF composite electrode [17] and heterogeneous catalysts for the Fenton reactions have been gaining attention in recent years. In several previous studies [18–21] it is demonstrated that the catalytic activity of the reaction system and the stability of the catalysts could be enhanced by binding iron species on appropriate support materials such as neutral organic polymers, ion exchange membranes or resins, and inorganic materials.

In the present study we want to demonstrate that iron can be effectively entrapped in a biopolymer matrix such as alginate without significant reduction in its reactivity. Entrapment within alginate beads is one of the most common methods for immobilizing living cells in food and beverage industries and also in the entrapment of surfactants, activated carbon, and metal hydroxides (Fe³⁺ and Ni²⁺) to recover/treat aqueous copper, organics, and arsenic. Immobilization in biopolymer matrix beads is a simple, inexpensive and effective technique. Thus, porosity of alginate beads allows solutes to diffuse into the beads and come in contact with the entrapped material. Moreover, they are nontoxic, biodegradable, and nonimmunogenic, producing thermally irreversible and water insoluble gels. In addition, another advantage of a supported catalyst system is the easy separation from the reaction solution by filtration, favoring the operation in continuous mode [22–24].

In this work, we report the development of an heterogeneous electro-Fenton catalysts treatment by means of Fe alginate gel beads in order to avoid the little activity for substrate degradation due to the precipitation of the Fe ions at pH >4 as well as difficult post-treatment of Fe sludge after the reaction. In order to analyse the technique efficiency, different dye solutions were used as model samples and comparative studies with respect to electro-Fenton with free Fe ions were carried out. Considering that the synthetic dyes present in the wastewater usually contain aromatic rings that make them harmful and biologically recalcitrant compounds, they represent a great environmental problem; the final aim of this work is to design an electro-Fenton process using Fe alginate gel beads to treat coloured effluents in continuous mode at bench scale.

2. Experimental

2.1. Dyes solutions

In this work two dye solutions (Lissamine Green B and Azure B) provided by Sigma–Aldrich (Barcelona, Spain) were used to evaluate the electro-Fenton treatment. The dyes characteristics and their concentrations are described in Table 1.

2.2. Fe alginate gel beads

Solution of sodium alginate 2.0% (w/v), purchased Sigma–Aldrich (Barcelona, Spain), was dropped into the hardening solution composed of 0.15 M Ba²⁺ (BaCl₂·2H₂O, Panreac Quimica, Barcelona, Spain) and 0.05 M Fe³⁺ (FeCl₃, Sigma–Aldrich (Barcelona, Spain) by a peristaltic pump and spherical alginate beads were formed [25]. These brown particles formed (Fig. 1A) were cured at 4 °C for 2 h in the gelling solution then filtered off and washed repeatedly with distilled water and finally stored at 4 °C in distilled water for the dye degradation study.

2.3. Characterization of Fe alginate gel beads

Scanning electron microscopy (SEM) was performed on a JEOL JSM-6700F equipped with an EDS Oxford Inca Energy 300 SEM using an accelerating voltage of 20 kV (Electron Microscopy Service, C.A.C.T.I., University of Vigo). The beads in the wet state were frozen in liquid nitrogen and freeze-dried. The freeze-dried beads were coated with C for the SEM observation.

Fourier-transform infrared (FT-IR) spectra of the beads were recorded on an FT-IR spectrometer (model FT-IR/4100, Jasco). The samples were ground into powder and dried in an oven at 60 °C for 40 min.

Fe alginate gel beads were digested to determine the Fe entrapped in the alginate beads following the EPA's acid digestion procedure 3050.

2.4. Electro-Fenton process

Batch electro-Fenton experiments were carried out in a glass cylindrical reactor with a working volume of 0.15 L (Fig. 2A). In this process H₂O₂ is produced electrochemically via oxygen reduction on the cathode. Therefore, continuous saturation of air at atmospheric pressure was ensured by bubbling compressed air near the cathode at about 1 L min⁻¹, starting 10 min before electrolysis to reach a stationary O₂ concentration [26].

The electric field was applied by two graphite sheets connected to a direct current power supply (HP model 3662). The electrode sheets (surface 15 cm²) were placed opposite to each other at 1 cm above the bottom of the cell and with an electrode gap of 6 cm. The current intensity was monitored along the process with a multimeter (Fluke 175). A constant potential drop (14.19 V) was applied with a power supply (HP model 3662) and the process was monitored with a multimeter (Fluke 175) [27].

In electro-Fenton with free Fe, the Fe dosage was initially added as FeSO₄·7H₂O at a concentration of 150 mg L⁻¹ and the pH was adjusted to working value, pH 2, with sodium hydroxide or sulphuric acid to promote the formation of hydroxyl radicals in the Fenton process [2]. When the electro-Fenton was performed with Fe alginate gel beads, 8.69 g of these beads were added in the solution. This amount of alginate beads have entrapped the same amount of Fe used when Fe is free. In these experiments pH was adjusted to working value with sodium hydroxide or sulphuric acid. In both cases, the solution was stirred magnetically avoiding concentration gradients in the cell.

2.5. Continuous electro-Fenton bubble reactor

A glass cylindrical reactor with two electrode sheets connected to a direct current power supply was used (Fig. 2B). The electro-Fenton bubble reactor had a working volume of 0.15 L, and 8.69 g of Fe alginate beads. It was operated in continuous mode at a residence time of 30 min. The cathode and anode graphite sheets (surface 15 cm²) were placed opposite to each other with an electrode gap of 4.3 cm. A constant potential drop (14.19 V) was applied with a power supply (HP model 3662) and the process was monitored with a multimeter (Fluke 175) [27].

As it is mentioned above, the H₂O₂ is produced electrochemically by bubbling compressed air near the cathode at about 1 L min⁻¹.

The hydrodynamic behavior of this reactor was studied through the residence time distribution (RTD) curves. The RTD curves were obtained by perturbation via a pulse injection with a syringe of 1 mL concentrated Lissamine Green B solution at the inlet of the bubble reactor without direct current. Then, the changes in dye concentration were measured along time in the outlet stream.

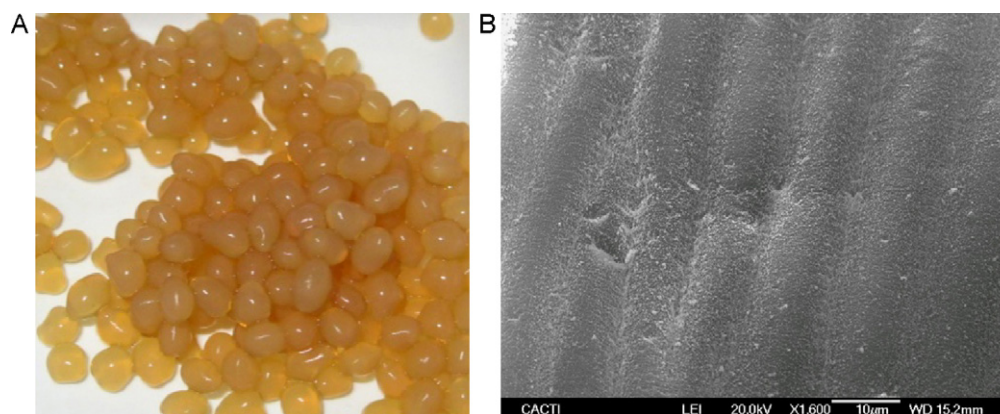


Fig. 1. The photographs of Fe alginate gel beads showing (A) the spherical shape of beads and their brown colour. (B) Scanning electron microscopy images of surface of Fe alginate beads.

2.6. Sample preparation

In all experiments, samples were taken periodically from the electro-Fenton cells and reactor to be analyzed for pH, dyes concentration and decolourisation, and electric parameters were recorded. They were centrifuged at 10,000 rpm for 5 min, and the supernatant was separated to be analyzed.

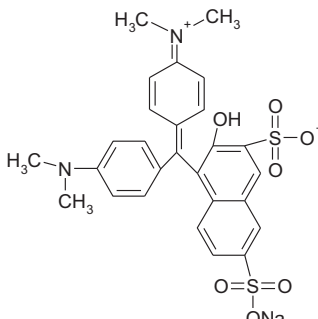
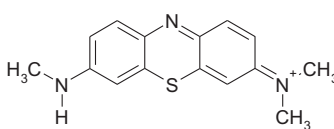
2.7. Dye decolourisation

The absorption spectra showed in all cases a single peak with a strong absorption in the visible region at the wavelength indicated in Table 1. For all experiments, the residual dye concentration was measured spectrophotometrically (Unicam Helios β , Thermo Electron Corp.) at the maximum visible wavelength. The assays were done twice, the experimental error was calculated as standard deviation (SD) and in all cases the SD was below 3%. Dye decolourisation was associated with the decrease in absorbance and expressed in terms of percentage according to Eq. (1).

$$D = \frac{A_i - A_t}{A_i} \times 100 \quad (1)$$

Table 1

Dye class, chemical structures, wavelength at maximum absorbance and concentration used of the different dyes employed.

Dye	Type	C.I.	Structure	λ_{\max} (nm)	Concentration (mg L ⁻¹)
Lissamine Green B	Diphenylnaphthyl-methane	44090		633	8.5
Azure B	Cationic metachromatic thiazine dye	52010		648	4.83

where D , decolourisation (%); A_i and A_t , absorbance value at the maximum wavelength of dyes at the initial and through time, respectively.

2.8. Kinetic studies

Kinetic studies were done in order to model the continuous electro-Fenton bubble reactor. The dyes concentration profiles were fitted by a suitable kinetic equation and the rate constants were calculated by using SigmaPlot 4.00 (1997) software. The SigmaPlot curve fitter uses an iterative procedure, based on the Marquardt–Levenberg algorithm, which seeks the values of the parameters that minimize the sum of the squared differences between the observed and predicted values of the dependent variable.

2.9. TOC analysis

Total organic content (TOC) was determined by using a Lange cuvette test (LCK 380) in a Hach Lange DR 2800. The sample was introduced in the Lange cuvette. Under the conditions of the test, the carbon forms carbon dioxide, which diffuses through a

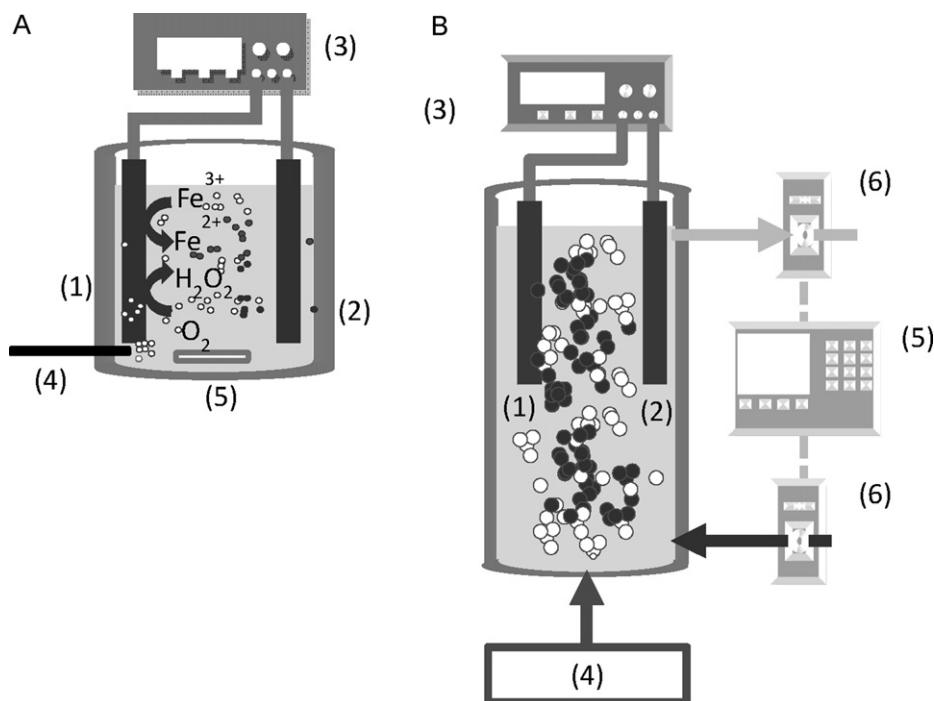


Fig. 2. Schematic diagram of electro-Fenton experimental setup in batch process A: cathode (1), anode (2), power supply (3), air (4), magnetic stirrer (5); and continuous process B: cathode (1), anode (2), power supply (3), air (4), pumps controller (5), pumps (6).

membrane into an indicator solution. The change of color of the indicator solution is evaluated photometrically.

3. Results and discussion

3.1. Characterization of Fe alginate gel beads

The Fe alginate beads were obtained via a traditional facile pathway. Their morphologies are shown in Fig. 1A. They exhibited spherical shape with a narrow particle size distribution and in the swollen state had average diameters of 3.5 mm. In comparison with alginate beads without Fe, it was observed that the colorless beads have transformed into brown, as a result of the cross-link between Fe and alginate.

It is well known that ionic cross-links will happen when alginate contacts with Ca²⁺ to form the “egg-box” structure. This structure is showed in the scanning electron microscopy images (Fig. 1B). Moreover, multivalent counter-ions as cross-linking molecules for the formation of three dimensional alginate gels have been reported [28,29]. Therefore, when alginate droplets are immersed

into ferrous aqueous solution, ionic cross-links can be established between the carboxyl group on alginate chains and Fe.

Using an acid digestion it was determined that the used Fe alginate beads have a Fe concentration of 2584 mg kg⁻¹. In order to determine the Fe distribution a SEM analysis was done. In Fig. 3, the SEM image and energy dispersive spectrometric (EDS) mapping of a Fe-alginate bead are showed. It is clear that the beads having spherical shape and the inner surface filled with iron are denser. The homogenous distribution of Fe in the beads revealed that alginate matrix was used successfully.

In addition, FT-IR spectra (Fig. 4) shows two bands at 1596.6 cm⁻¹ (III) and 1406.2 cm⁻¹ (II), which are assigned to the asymmetric and symmetric stretching vibrations of the carboxyl group of alginate molecule, respectively [30,31]. According to Dong et al. [32], a new peak around 1710–1720 cm⁻¹ (IV) in the spectrum of Fe alginate gel bead may also belong to the absorption vibrations of the carboxyl group of alginate molecule. Moreover, it is observed that peak II and III shifted slight to 1595.0 cm⁻¹ and 1407.1 cm⁻¹ in the spectra of Fe alginate gel bead. This fact is indicative of the corresponding change in the distance of the C–O bond of the

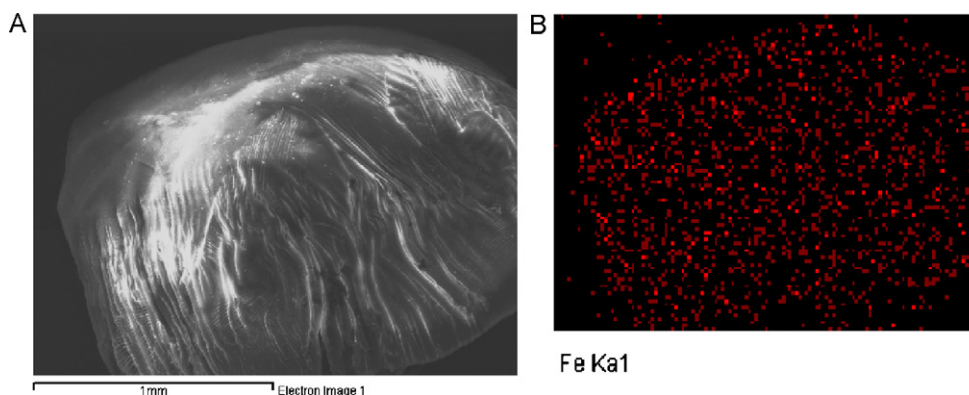


Fig. 3. Scanning electron microscopy images (A) and energy dispersive spectrometric mapping of a Fe alginate gel beads (B).

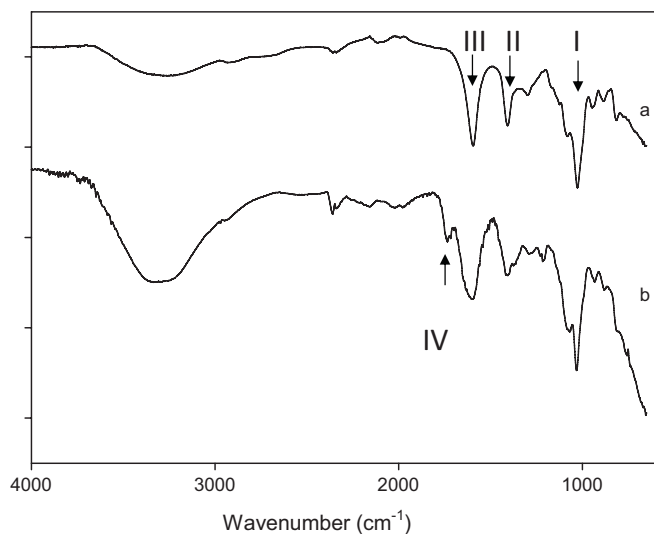


Fig. 4. Infrared spectra of: (a) sodium alginate and (b) Fe alginate bead.

carboxylate group is due to the bonding strength between the metal ion and the oxygen of the carboxyl group changes. Hence, the bond needs a smaller amount of energy to vibrate; therefore, a shift of the asymmetric peak to lower energies is observed [31]. These results could indicate that a surface gelation of sodium alginate can be formed by the complex of their carboxyl groups with iron ions in aqueous solution.

3.2. Dye adsorption on alginate beads

As it is mentioned in the literature [33–35], several polymer network gel beads could be used in the removal of inorganic and organic contaminants present in wastewater. Jeon et al. [34] examined the removal of some basic dyes, such as Methylene Blue, Malachite Green and Methyl Orange, using alginate or alginate/polyaspartate composite gel beads. Their results suggest that this gel can be used as an effective sorbent for water pollutants such as dyes. Li et al. [35] reported that chitin/alginate magnetic nano-gel beads possessed high Methyl Orange adsorption capacity due to the increase of the specific surface area for composite beads because of the microporous structure and the formation of iron oxide nanoparticles, which can adsorb more easily dye molecules. Therefore, the iron oxide nanoparticles of composite beads play an important role in the improvement of both the adsorption capacity and the adsorption rate.

Considering that the utilized matrix could be used as sorbent, initial adsorption experiments to evaluate the contact time effect on dye removal by beads were carried out at dye concentration of Table 1 and room temperature. The results obtained at the dye concentrations used in these experiments showed that a dye adsorption lower than 5% is attained when the beads are in contact more than 1 h. Therefore, if the electro-Fenton process takes place in lesser time the effect of adsorption on alginate bead could be considered as null.

3.3. Effect of solution pH

In this work the electro-Fenton process was accomplished in an electrochemical cell with graphite electrode based on previous studies [2,36]. Graphite was the best cathode material for electro-generation of H_2O_2 while metal cathodes such as copper, stainless steel, lead and nickel were likely to decompose H_2O_2 . Additionally, this electrode permits an adequate H_2O_2 production according to the necessary ratio $Fe:H_2O_2$ in order to Fenton reaction take

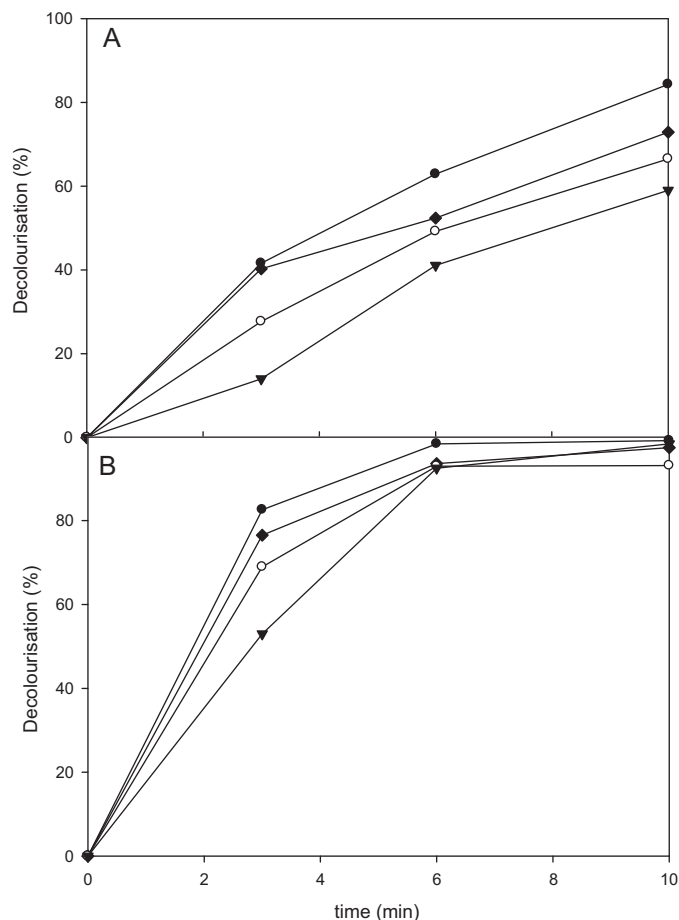


Fig. 5. Effect of pH on decolourisation of Azure B (A) and Lissamine Green B (B) by electro-Fenton with Fe alginate gel beads (8.69 g) with a working volume of 0.15 L, air flow 1 L min^{-1} , potential drop 14.19 V . Symbols: pH 2 (●), pH 4 (◆), pH 6 (○), pH 8 (▽).

place [37]. Amongst the different pollutants that provoke a colour environmental problem, Lissamine Green B and Azure B dyes were selected as model dyes because they are recalcitrant compounds which are not easily oxidized by other methods [38]. Therefore, in this study the Lissamine Green B and Azure B decolourisation by electro-Fenton technique by use of catalytic activity of Fe alginate gel beads was carried out in an electrochemical cell with graphite sheet electrodes. The concentration range of Azure B and Lissamine Green B was selected in accordance with the levels used in previous papers [39–41].

The efficiency of the Fenton reactions depends on pH, for that reason its impact on the catalytic activity of Fe alginate gel bead was examined and several experiments were performed at pH 2, 4, 6 and 8 in dye solutions containing 8.69 g gel beads (Fig. 5).

As it is shown in Fig. 5, solution pH influences dye decolourisation and the highest oxidation activity is achieved at pH 2. This agrees with the optimal value for the Fenton's reagent reported by Rosales et al. [2]. Decolourisation exhibits different rates for the pH range used. In the initial stages, dye decolourisation rate at pH 2 is much faster than that obtained with higher pH, while in the final stage the percentage of degradation is similar for all tested conditions. This trend is mostly due to the fact that at lower pH values the scavenging effect of the OH radicals by H^+ is severe (Eq. (2)) [42] and H_2O_2 is unstable in alkaline solutions, being quickly decomposed to oxygen and water at neutral to higher pH values, which leads to lose its oxidation ability (Eq. (3)) [43].



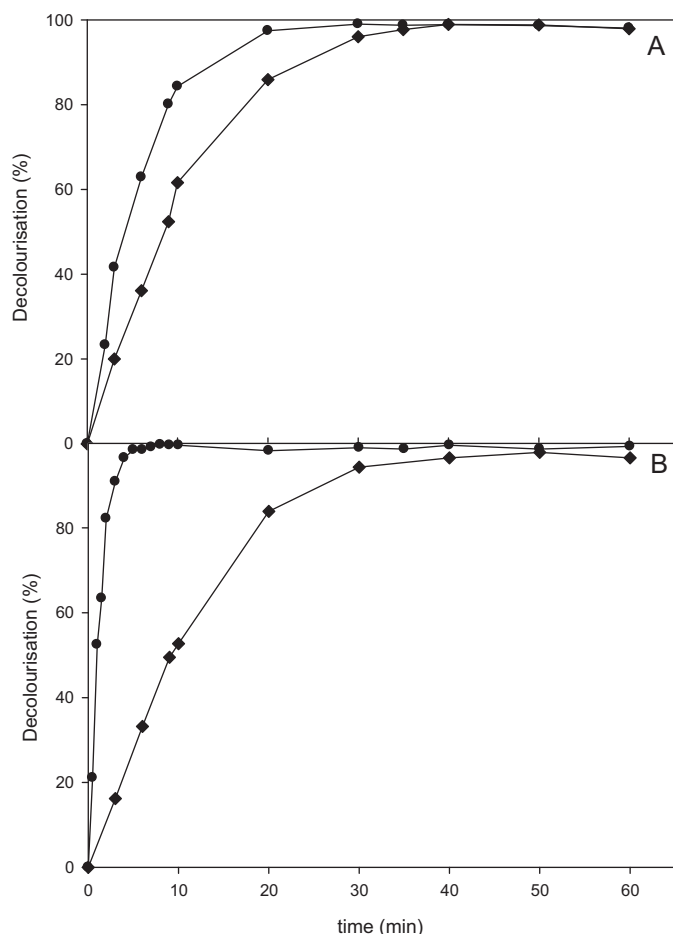


Fig. 6. Electro-Fenton treatment of Azure B (A), Lissamine Green B (B) using free Fe (♦) and Fe alginate beads (●), operating in a electrochemical cell with a working volume of 0.15 L, air flow 1 L min⁻¹, potential drop 14.19 V and pH 2.



However, in electro-Fenton the continuous H₂O₂ production permits to maintain their concentration in the medium. For this reason, at initial time the decolourisation efficiency was reduced but at last time the removal level was the same as pH 2. Moreover, the ionic cross-links between the carboxyl group on alginate chains and Fe ions avoid the formation of Fe(OH)₃ and eliminate the negative effect of iron sludge generated in classical Fenton's oxidation process at higher pH values. The results demonstrated that Fe alginate beads played a great catalytic role in the decolourisation of dyes in a wide pH range of 2–8.

3.4. Comparative electro-Fenton process operation with Fe alginate beads and free Fe

The decolourisation of Lissamine Green B and Azure B under electro-Fenton process with free and immobilized Fe ions was tested in the electrochemical cell described in the material and methods section. A pH 2 was selected due to a previous study [2] it was detected that with free Fe decolourisation efficiency significantly decreases, mainly because Fe³⁺ precipitates at high pH values, and the dissolved iron concentration is reduced.

As it can be observed in Fig. 6, the oxidation of both dyes performed in the presence of Fe alginate beads increased the reaction rate significantly, and high colour removals (superior than 95%) were reached in shorter treatment time. After experiments, negligible Lissamine Green B and Azure B dye presence were detected in

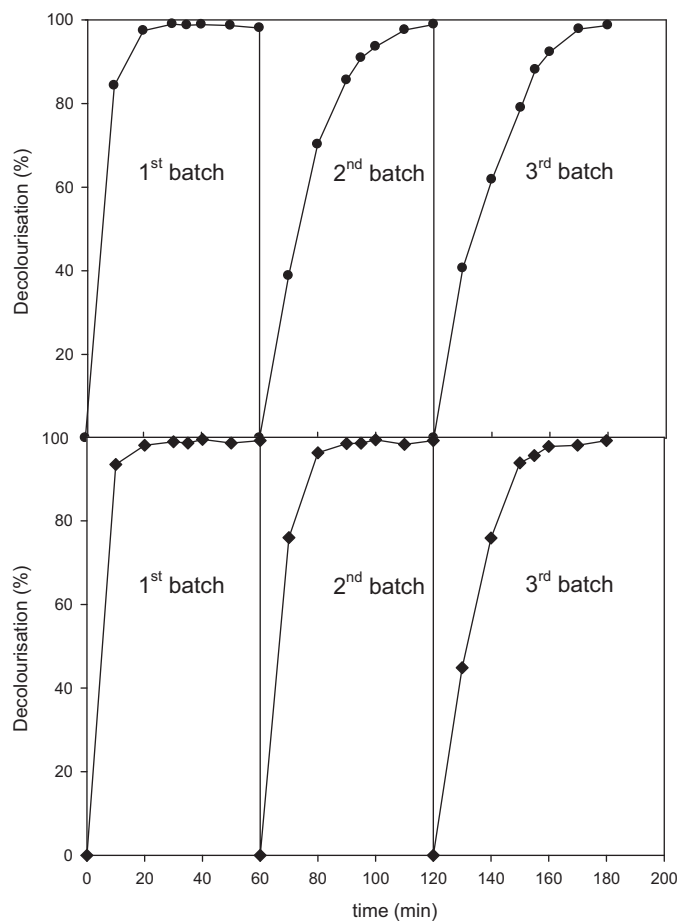


Fig. 7. Dye decolourisation in successive electro-Fenton batches with 8.69 g of Fe alginate beads operating with a working volume of 0.15 L, air flow 1 L min⁻¹, potential drop 14.19 V and pH 2. Symbols: Azure B (●), Lissamine Green B (♦).

the electrodes and Fe alginate beads. Therefore, these results clearly indicate that both dyes were decolourised and also degraded during electro-Fenton process with the heterogeneous Fenton catalysts designed in this study.

3.5. Successive batches

Successive batches were performed to examine the reusability of the Fe alginate beads for dye decolourisation. As it can be observed in Fig. 7, after 3 cycles, the time necessary to obtain the maximum dye decolourisation was increased. Although after 1 h the decolourisation level attained remained over 98–100% for the 2 and 3 cycles, this value was obtained in the first batch after 30 min. In order to evaluate the fixation of Fe in the alginate beads, a SEM image and energy dispersive spectrometric (EDS) mapping of a Fe alginate bead were done. It is clear that the beads maintain their spherical shape and are filled with Fe (Fig. 8).

Thus, alginate Fe immobilization appeared to exhibit a high reusability with high stability and the homogenous distribution of Fe in the beads permit the electro-Fenton process to take place in successive batches without addition of Fenton agents.

3.6. Continuous electro-Fenton process

Previous results demonstrated that the use of Fe alginate gel beads permits their reuse in successive batches. Thus the continuous mode operation was tested in a bubble reactor. Initially the hydrodynamic behavior of this reactor was studied through

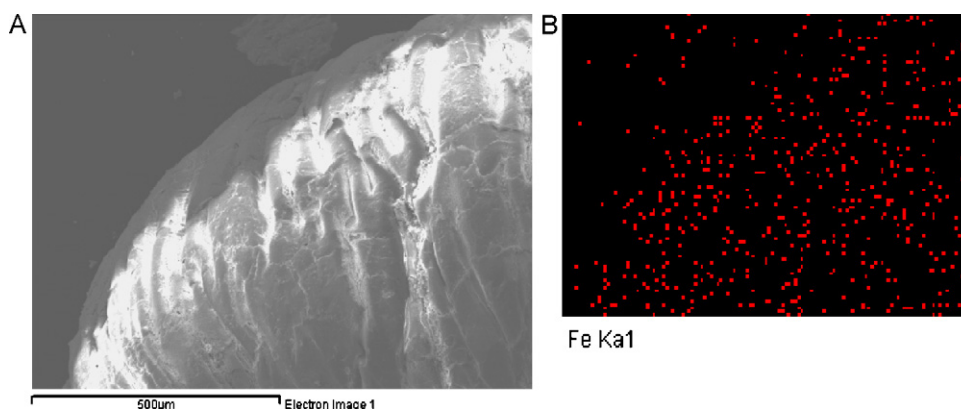


Fig. 8. Scanning electron microscopy images (A) and Energy dispersive spectrometric mapping (B) of Fe alginate after 3 batch uses.

the residence time distribution (RTD) curves. These tracer experiments were carried out with 8.69 g of Fe alginate beads at different flow rates to confirm that the reactor used in this work closely matches ideal mixing conditions. Therefore, it can be assumed that the reactor behaves CSTR ideally, which means that the concentration everywhere in the reactor is equal to the outlet concentration and the fluid has a mean residence time equal to the reactor volume divided by the volumetric flow rate through the tank.

To model this process is necessary to know its kinetic behaviour in the previous batch experiments. For this reason, the kinetic studies with both dyes were carried out. In these experiments the reaction kinetics were researched and the regression coefficients for zero, first- and second-order reactions were calculated. The results indicated that the decolourisation of Lissamine Green B and Azure B could be quantitatively described by a first-order kinetic equation (Eq. (4)) with respect to the dye concentration:

$$\frac{dC}{dt} = -kC \quad (4)$$

where C , concentration of dye (mg L^{-1}); t , reaction time (min); k , kinetic coefficient for the first order reaction (min^{-1}). The rate constant values and the statistical correlation parameters are shown in Fig. 9.

Finally, the efficiency of the continuous bubble electro-Fenton reactor designed in this work was evaluated operating with Lissamine Green B and Azure B at a residence time of 30 min. Fig. 10 shows the decolourisation efficiency to Lissamine Green B and Azure B. As it is shown, both dyes decolourisation profiles were very similar, around 98–100% of dye decolourisation can be obtained operating in continuous mode without operational problems, such as the clogging, bead breakage, or overpressure. To verify the electro-Fenton dye degradation, the reduction in TOC was evaluated in both dyes. TOC removal was higher by Lissamine Green (93%) than Azure B (89%). The results demonstrated that colour removal is closely related with the reduction of TOC and these results are in accordance with the one obtained in our previous experiments [2] and by Daneshvar et al. [44]. They reported that the electro-Fenton process can degrade and finally mineralize organic pollutants to less toxic and non-toxic compounds. By reacting with hydroxyl radicals, the dye is degraded step-by-step, and eventually mineralized.

Based on the kinetic and hydrodynamic studies it was postulated the equation of an ideal CSTR in steady-state (Eq. (5)).

$$D = \frac{k \cdot \tau}{1 + k \cdot \tau} \times 100 \quad (5)$$

where, D , decolourisation of dye (%); τ , residence time (min) and k , kinetic coefficient for the first order reaction (min^{-1}). In Fig. 10 the line represents the model and points the experimental data. It

clearly shows a good fit ($R^2 \cong 0.99$) to the theoretical model and verifies the utility of this model to scale-up the process.

These results suggest that the oxidative degradation of dyes under electro-Fenton process using Fe alginate gel beads in continuous reactor can be achieved. In this case, the alginate beads are easily retained into de reactor and different kinds of dyes were treated without operational reactor problems and attaining the dye decolourisation without the addition of other oxidant agents.

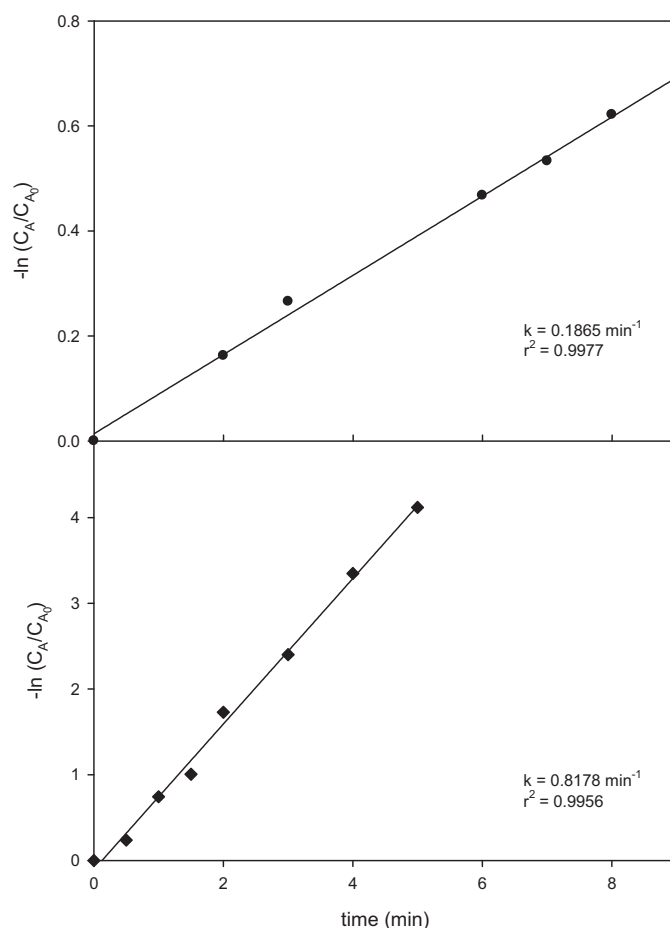


Fig. 9. First-order kinetics (Ln normalized dye concentration remaining vs time) of dye decolourisation by electro-Fenton process (8.69 g of Fe alginate beads, working volume of 0.15 L, air flow 1 L min^{-1} , potential drop 14.19 V and pH 2). Symbols: Azure B (●), Lissamine Green B (◆).

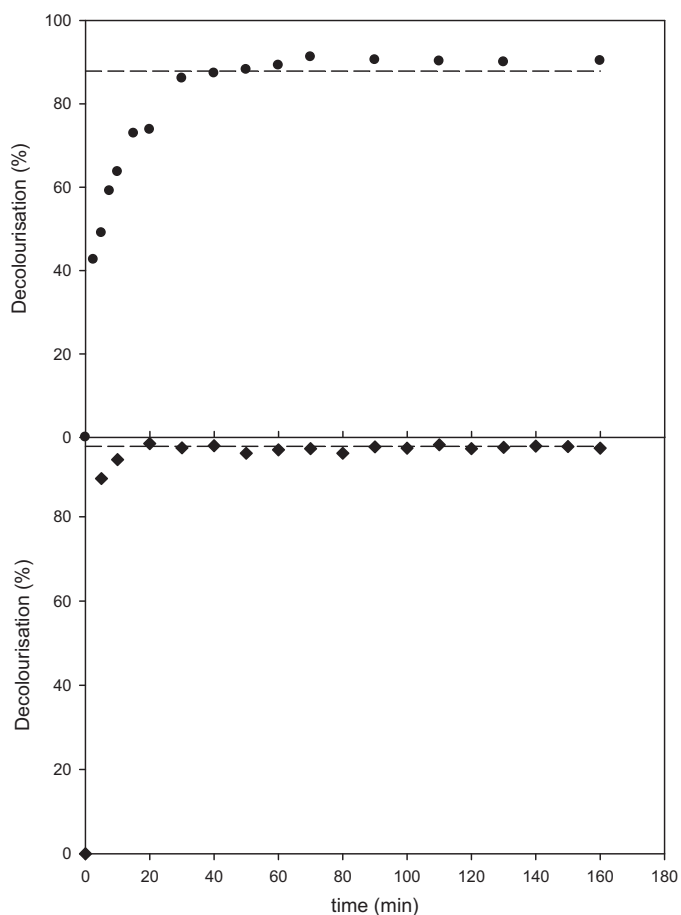


Fig. 10. Continuous electro-Fenton treatment of Azure B (●) and Lissamine Green B (◆) in a bubble reactor with a residence time of 30 min (8.69 g of Fe alginate beads, working volume of 0.15 L, air flow 1 L min⁻¹, potential drop 14.19 V and pH 2). Line represents the model prediction.

4. Conclusions

In this study a new variation of electro-Fenton process by using of catalytic activity of Fe alginate gel beads has been developed. The performance of this technique is described by:

- The characterization of Fe alginate gel beads revealed that Fe is homogeneously distributed into the gel beads.
- The Fe fixation into alginate beads reduces the negative effect of pH in the electro-Fenton process. It is possible to operate in a wide range of solution pH (from 2 to 8).
- The catalytic activity of Fe alginate gel beads improved the dye decolourisation of electro-Fenton technique.
- After successive batches, the stability of Fe alginate gel beads was maintained and Fe was fixed in the alginate structure.
- An oxidative degradation of dyes under electro-Fenton process using Fe alginate gel beads in continuous reactor can be achieved.
- A theoretical model was proposed in accordance with experimental data and the utility of this model to scale-up of the process was verified.

The results obtained in this study, indicate the suitability of electro-Fenton technique in presence of Fe alginate gel beads to treat colored effluents that can solve one of the most important environmental problems in the related industry.

Acknowledgements

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