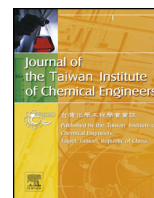




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Fe-modified local clay as effective and reusable heterogeneous photo-Fenton catalyst for the decolorization of Acid Green 25

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ABSTRACT

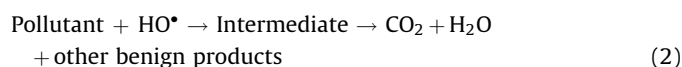
Locally sourced Bandar Baru clay (BBC) from Kedah in Malaysia was modified with NaCO₃ and applied as catalyst support. Six different Fe supported BBC (Fe-BBC) catalysts were prepared with different calcination temperature (300–500 °C) and time (4–6 h) and their activities were tested on the decolorization of Acid Green 25 (AG25) pollutant dye. The best Fe-BBC was that calcined at 300 °C for 4 h, and it was characterized. The BET result showed increase in surface area and pore volume which was supported by the SEM result. The presence of the incorporated Fe in the catalyst was verified with EDX, XRD and FTIR techniques. The best experimental condition decolorized 99% of 50 ppm (AG25) in 20 min with 1.25 g/L Fe-BBC300/4 and 6.7 mM of hydrogen peroxide at pH 3. Its reusability after three cycles showed 97.5% decolorization of 50 ppm of AG25. The versatility of Fe-BBC300/4 was also verified on the decolorization of Reactive Blue 4 and Direct Blue 71.

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1. Introduction

Currently the world technological development ranging from industrial, pharmaceutical and other allied areas such as military weapon development and advance research into newer products are on the highest ever increasing rate. Consequently, the proliferation of different conventional and alien pollutants is also on the alarming rate of increase. Current researches toward curtailing and minimizing the effects of these deleterious pollutants on the environment is also gathering momentum especially in the fields of applied chemistry and environmental engineering bearing in mind both the economics of the researched processes and their sustainability. Different methods of pollutant degradation and mineralization such as biological, physical and chemical methods have been widely studied and reported [1–5]. These methods can be used separately or combined to enhance the overall treatment efficiency [1]. The advent of Fenton process which is an advance oxidation process (AOP) where ferrous ion (Fe²⁺) is employed to generate hydroxyl radical (HO•) with sufficiently high oxidation potential of 2.8 eV from hydrogen peroxide (HP) (Eq. (1)) had made easy the degradation, decolorization and even mineralization of virtually all known pollutants at ambient conditions [2–4]. After each cycle of

HO• generation (Eq. (1)) and subsequent usage for pollutants depuration (Eq. (2)), the Fe²⁺ is oxidized to Fe³⁺ (Eq. (1)) which must be reduced to Fe²⁺ (Eq. (3)) for another cycle of reaction. A more economical and efficient reduction process for Fe³⁺ known as photo-reduction process is depicted by Eq. (4), which has been reported to be superior to the route shown in Eq. (3) [2,5], the overall process is referred to as photo-Fenton process [1–8]. In addition, the required HO• can also be generated from HP according to Eq. (5), a procedure referred to as photolysis of HP [5].



Conventionally, photo-Fenton process had been practiced in the homogeneous phase but the overall efficiency of the process is considered to be generally poor due to the slow rate of Fe²⁺ regeneration from Fe³⁺ [9] and inability to easily recover the Fe catalyst due to the formation of ferric sludge which makes the process require additional processing steps like coagulation, sedimentation and filtration [6]. In addition, the process can only

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be operated within a narrow pH range [5,10]. In view of the above, the replacement of the homogeneous photo-Fenton process with heterogeneous where the active metal can be conveniently intercalated into solid supports becomes an essential alternative. Several attempts have been made to develop supported catalysts using rice husks, graphite and activated carbon [11–13]. Recently, we also reported the application of industrial processed clay such as kaolin, montmorillonite and bentonite in the degradation of different pollutants in photo-Fenton [1,2,13,14] and Fenton-like process [5]. All these clay supports and several others reported in some critically reviewed articles [4,8,15] were purchased as industrial processed clay minerals which increases the cost of catalyst development and in turn challenged the industrial application of heterogeneous photo-Fenton process. In order to make this process economically attractive, the need to substitute these industrially refined supports with low cost and locally sourced materials such as raw clay becomes expedient.

In this report, raw clay from the paddy field at Bandar Baru, Kedah, Malaysia was modified with Na_2CO_3 and studied as heterogeneous catalysts support for photo-Fenton process. The catalysts were screened for the best calcination temperature and time on the decolorization of Acid Green 25. The best catalyst was characterized for physical and chemical properties, tested at different operational conditions and its versatility was validated on the decolorization of Reactive Blue 4, Acid Green 25 and Direct Blue 71.

2. Material and methods

2.1. Catalyst support preparation

The catalyst support is Bandar Baru clay (BBC) obtained from the paddy field at Bandar Baru in Kedar, Malaysia. Prior to its use, the gray colored BBC was grinded to powder using laboratory mortar and washed several times with double distilled water to remove impurities. The samples were dried in an oven at $100\text{ }^\circ\text{C}$ for 12 h followed by sieving to obtain particle size range of 63–75 μm .

2.2. Catalyst preparation

The BBC supported Fe (Fe-BBC) Fenton catalyst was prepared by impregnation method where 0.2 M of $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ obtained from Merck Co. was dissolved in a beaker containing distilled water and vigorously stirred. Na_2CO_3 was added to obtain a molar ratio of 1:1 ($\text{Na}^+/\text{Fe}^{3+}$) in order to enhance the possibility of successful intercalation of the Fe^{3+} into the BBC support via simple ion exchange reaction where the Na^+ can be replaced by Fe^{3+} [16,17]. In addition, it had been reported that the addition of Na_2CO_3 at the catalyst synthesis stage is capable of increasing both the surface area and pore volume [16–18]. 2.0 g of the powdered BBC was added to the $\text{Na}^+/\text{Fe}^{3+}$ solution and was continuously stirred for 2 h followed by drying in an oven at $80\text{ }^\circ\text{C}$ for 24 h. The dried solid was washed several times with double distilled water to remove excess Na^+ ions and re-dried in an oven at $80\text{ }^\circ\text{C}$ for 12 h [16,19]. Finally, the obtained sample was calcined at temperature and time range of 300–500 $^\circ\text{C}$ and 4–6 h, respectively in a muffle furnace and labeled for instance as Fe-BBC300/4, for Fe-BBC sample calcined at 300 $^\circ\text{C}$ for 4 h.

2.3. Catalyst characterization

The catalysts were characterized by nitrogen adsorption/desorption isotherm using Brunauer–Emmett–Teller (BET) method, energy dispersive X-ray (EDX), Fourier transformed infrared spectroscopy (FTIR), X-ray diffraction (XRD) and scanning electron microscope (SEM).

Nitrogen adsorption–desorption measurements with BET method were performed at liquid nitrogen temperature ($-196\text{ }^\circ\text{C}$) with an autosorb BET apparatus, Micromeritics ASAP 2020, surface area and porosity analyzer. The analysis procedure is automated and operates with the static volumetric technique. Before each measurement, the samples were first degassed at $200\text{ }^\circ\text{C}$ for 2 h. EDX was performed to determine the chemical composition in the samples using the same instrument with the SEM. FTIR analyses were performed on the samples, using a Perkin-Elmer Spectrum GX Infrared Spectrometer with resolution of 4 cm^{-1} , in the range of $4000\text{--}400\text{ cm}^{-1}$. The sample and analytical grade KBr were dried at $100\text{ }^\circ\text{C}$ over-night prior to the FTIR analysis and were prepared with the disc technique using a finely ground mixture of 0.25 mg of sample and 100 mg of KBr. The XRD patterns of the samples were measured with Siemens XRD D5000 equipped with $\text{Cu K}\alpha$ radiation and recorded in the range of $5\text{--}90^\circ$ with a scanning rate of 2° min^{-1} . SEM was used to study their surface morphology, the analysis was carried out using a scanning electron microscope (Model EMJEOL-JSM6301-F) with an Oxford INCA/ENERGY-350 microanalysis system.

2.4. Experimental procedure

All experiments were carried out in 1 L beaker filled with 200 mL of AG25 dye solutions with initial concentration of 50 ppm (except for studies on the effect of initial dye concentration). The beaker was placed on a hot thermostated variable speed magnetic stirrer. The temperature and pH was monitored using EcoScan SC11-4115 (EXATECH ENTERPRISES) probe inserted inside the beaker. For each experimental run, the solution pH was adjusted with the addition of 1.0 M H_2SO_4 or 1.0 M NaOH followed by the addition of catalyst. The reaction was initiated immediately after the UV irradiation lamp (254 nm, 0.75 W/m^2 , model GPX9 9W, Jiangyin Juguang Photoelectric Instrument Co., Ltd., China) was switched on and the desired amount of HP added. The extent of decolorization was monitored by taken filtered samples from the reaction mixture at selected time intervals and measured using UV–vis spectrophotometer. Similar procedure was repeated for the study on the versatility of the catalyst to decolorize other pollutant dyes namely Reactive Blue 4 (RB4) and Direct Blue 71 (DB71).

2.5. Analytical methods

The maximum absorbance wavelength (λ_{max}) of AG25, RB4 and DB17 recorded from 700 to 200 nm using a spectrophotometric quartz cell in a UV–vis spectrophotometer (Shimadzu, model UV 1700 PharmaSpec, Japan) was found at 642, 598 and 584 nm, respectively. The withdrawn filtered samples were quickly analyzed at the established wavelength to minimize experimental errors since the reaction could still continue after withdrawal. The analyzed samples were again quickly re-analyzed three times for consistency and the average values were recorded. The decolorization efficiency of the dye pollutants was evaluated as follows:

$$\text{Decolorization efficiency (DcE)} = \left[\frac{C_0 - C_t}{C_t} \right] \times 100\% \quad (6)$$

where C_0 and C_t are the initial concentration and the measured concentration of the pollutants at the time of withdrawal, respectively.

2.6. Catalyst reusability test

For stability test, the catalysts were tested in three different consecutive experiments using fresh dye solution at the best observed experimental conditions. After each experimental runs, the used catalyst was filtered, washed with distilled water, and

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