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THE INFLUENCE OF Fe CONTENT IN Al, Fe-PILLARED CLAYS ON ITS PERFORMANCE IN CATALYTIC WET OXIDATION

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Abstract

In this work a series of Al,Fe-pillared clays (PILCs) with different Fe^{3+} content was synthesized and characterized. Their catalytic performance was studied in the in catalytic wet peroxide oxidation (CWPO) of food dye tartrazine used as a model compound. Degree of decolorization of tartrazine containing aqueous solution was monitored in relation to Fe^{3+} content. The results of the catalytic tests showed that higher Fe content leads to higher catalyst efficiency in the investigated reaction. The reaction can be described by 0 order kinetics inherent to heterogeneous catalytic processes.

Introduction

Al,Fe-pillared clays (PILCs) have shown good performance in catalytic wet peroxide oxidation (CWPO) of organic pollutants in water. They have predominantly been tested in the degradation of phenol [1]. There are seldom reports on their use in the degradation of other organic pollutants such as toluene [2] and dyes [3]. In this paper local bentonite clay (Bogovina) rich in smectite was used to obtain Al,Fe-PILC with different Fe^{3+} content. Their catalytic performance was studied in the in CWPO of tartrazine dye at moderate temperature.

Experimental

Bentonite was obtained from Bogovina, Serbia. It was crushed, ground and sieved through a 74 μm sieve and by hydroseparation a fine particle fraction < 2 μm was obtained. The PILCs were obtained using a common procedure [4] consisting of the following steps: grinding, sieving, Na exchange, intercalation, drying and calcination. Fe^{3+} to $(Al^{3+}+Fe^{3+})$ molar ratios in the pillaring solutions were 0%, 1%, 5%, 10% and 15% and the corresponding materials were denominated Al–PILC, Al,Fe1–PILC, Al,Fe5–PILC, Al,Fe10–PILC and Al,Fe15–PILC, respectively. The chemicals used for the Na^+ exchange and pillaring were NaCl, NaOH and Al(III) and Fe(III) nitrate.

X-Ray diffraction (Philips PW 1710 X-ray powder diffractometer with a Cu anode) confirmed that the pillaring was successful [5]. Chemical composition of the PILCs was determined using Spectro Spectroflame M - inductively coupled plasma optical emission spectrometer. Catalytic tests were carried out in a semibatch reactor under stirring and constant temperature maintained by circulation of thermostatic fluid using Julabo MC 4 heating circulator. Initial dye concentration was 50 ppm in the presence of excess of H_2O_2. Catalytic performance of the catalysts was examined using UV-Vis spectrophotometry.

Results and Discussion

The results of chemical analysis for the most abundant cations are given in Table 1.

Table 1. Chemical composition of samples

Sample	Si	Al	Fe
	mmol/100g of sample		
Na -B	910.3	474.3	107.3
Al PILC	916.5	859.3	110.9
Al,Fe-1 PILC	912.4	740.7	132.4
Al,Fe-5 PILC	921.3	715.7	178.9
Al,Fe-10 PILC	911.6	656.4	214.7
Al,Fe-15 PILC	925.5	637.0	304.1

Chemical analysis showed that Al pillaring lead to abrupt increase of the Al^{3+} content, while the inclusion of Fe^{3+} in the pillaring solution lead to gradual increase of the Fe content followed by decrease in the Al content of the obtained materials. These results are in good correlation with the Fe^{3+}/Al^{3+} ratios applied in the syntheses.

Fig. 1 represents dependence of tartrazine concentration (C_{tart}) on the reaction time during CWPO at 35 °C and shows the influence of Fe^{3+} content on the kinetics of dye degradation. The dependence acquires linear trend soon after the initiation of the reaction. Therefore, here presented data do not include 0 min point corresponding to the initial dye concentration.

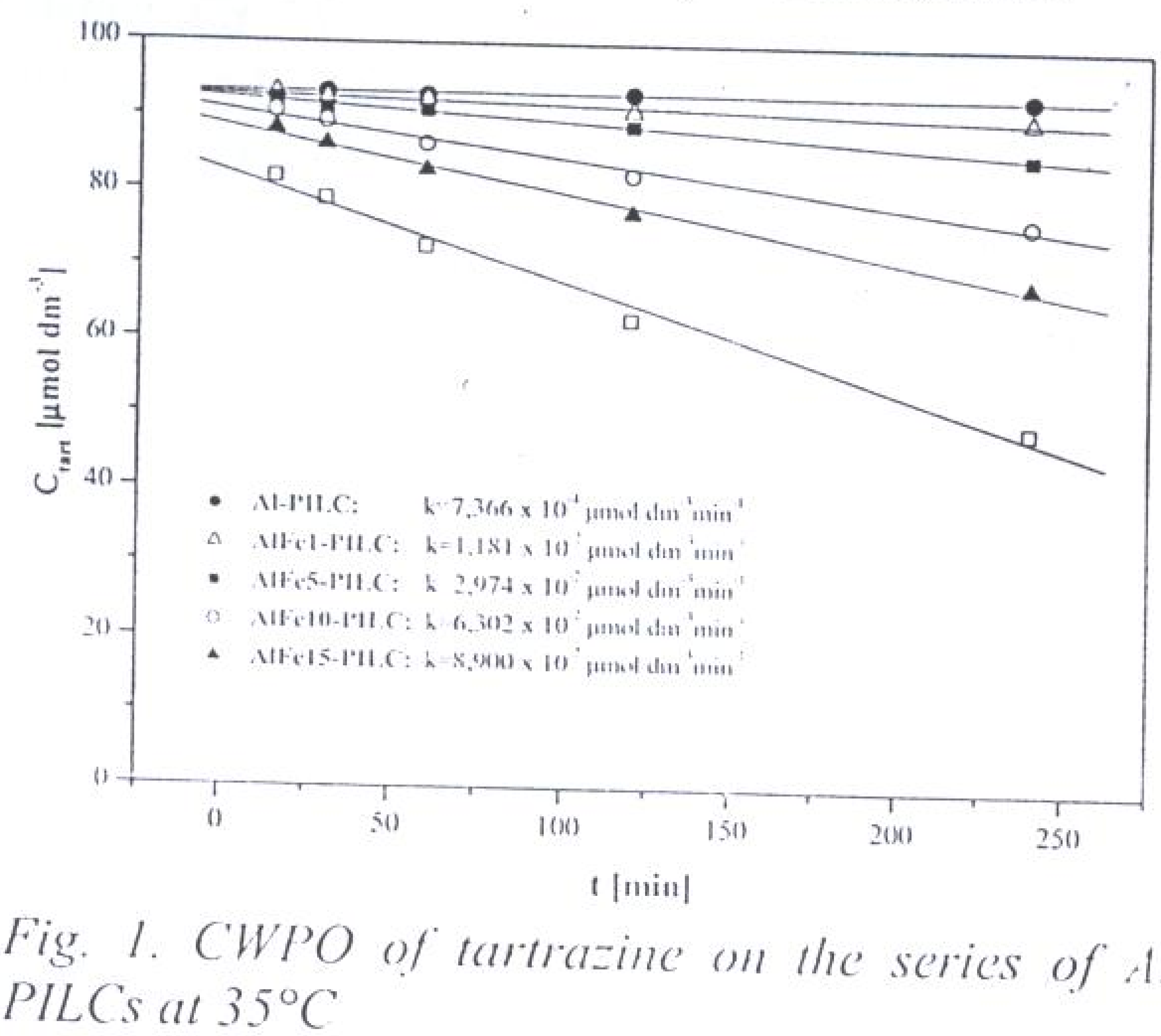

Fig. 1. CWPO of tartrazine on the series of Al,Fe–PILCs at 35°C

Linear dependence of $C_{tart}=f(t)$ indicates 0 order reaction kinetics established when adsorption/desorption equilibrium at active sites of the catalyst is reached. At that moment the reaction rate becomes independent on the C_{tart} in the solution. Higher Fe content of the catalysts means greater active site concentration resulting in greater reaction rate constant.

Although the investigated method did not involve pH adjustments, the catalytic tests showed good results even at mild temperature. Performance of the obtained Al,Fe–PILCs in the degradation of tartrazine was good. It was found to be dependant on the Fe^{3+} content which was a proof of the activity of incorporated Fe species in the investigated CWPO process.

Conclusion

A series of Al,Fe-pillared clay (PILC) materials with different Fe content was synthesized. The obtained materials were tested in the catalytic wet peroxide oxidation (CWPO) of azo dye tartazine at mild conditions. It was found that the Al,Fe-PILCs show activity in the dye degradation. The results of the catalytic tests showed that higher Fe content leads to higher catalyst efficiency in the investigated reaction. The reaction can be described by 0 order kinetics inherent to heterogeneous catalytic processes. Here investigated method was proven to be efficient in the decolorization of tartrazine containing water, thus reducing sunlight cut-off effects, and is prospective as a first step in a two stage water purification method, where the second one could be biodegradation.

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