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Fundamental and Applied Aspects of
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CATALYTIC OXIDATION OF TARTRAZINE IN THE PRESENCE OF RADICALS GENERATED FROM POTASSIUM PEROXYMONOSULFATE USING COBALT IMPREGNATED PILLARED MONTMORILLONITE

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ABSTRACT

In this work aluminum pillared clay impregnated with cobalt (CoAP) was tested as catalyst in the degradation of azo-dye tartrazine in the presence of potassium peroxymonosulfate (Oxone®). Aluminum pillared clay was synthesized using $\leq 74 \mu\text{m}$ fraction of Na-exchanged clay from Wyoming, USA (Na-Wy) using common procedure. The influence of Oxon®/catalyst ratio on catalytic performance was investigated. The cobalt impregnated pillared montmorillonite was found to be promising catalyst for dye decolorization.

INTRODUCTION

Potassium peroxymonosulfate (Oxone®, $\text{KHSO}_5 \cdot 0.5\text{KHSO}_4 \cdot 0.5\text{K}_2\text{SO}_4$) has increasingly been gaining attention as effective substitute for Fenton's reagent in the catalytic oxidation of water pollutants [1]. Oxone® is commercially available and environmentally-friendly source of $\text{SO}_4^{\cdot-}$ radicals that have similar or even higher oxidation potential than OH^{\cdot} , higher selectivity for unsaturated bonds and aromatic constituents and relatively long half-life comparing with that of OH^{\cdot} [2]. Activation of Oxone® is necessary in order to generate $\text{SO}_4^{\cdot-}$ radicals [3]. Oxone® is the most effectively activated by transition metallic cations, with cobalt showing the best performance [4]. Several studies have proposed homogeneous cobalt ions as a catalyst for decomposition into radicals [5]. The difficulty of recovering cobalt ions for preventing possible subsequent pollutions led to attempts to make the catalyst where cobalt is immobilized onto various supports [3].

In this work cobalt impregnated pillared montmorillonite was used as a catalyst for Oxone® “activation” in catalytic oxidation of tartrazine dye as the water pollutant.

EXPERIMENTAL

The $\leq 74 \mu\text{m}$ fraction of clay from Wyoming, USA rich in montmorillonite, was used for pillaring. After Na-exchange (Na-Wy) the sample was modified by pillaring. The process of pillaring was carried out according to a common procedure comprising the following steps: pillaring, rinsing, drying and calcination [6]. The obtained pillared clay was impregnated with cobalt using incipient wetness impregnation method and denoted as CoAP.

Tartrazine degradation by Oxone® without catalyst as well as catalytic tests with Na-Wy and CoAP were conducted. Catalytic tests were performed in a system containing potassium peroxymonosulfate (Oxone®) as the source of peroxy radicals and aqueous solution of tartrazine as model pollutant. The concentration of Oxone® and the mass of the catalyst were varied. Decolorization was monitored using Thermo Scientific, Evolution 220 UV–Visible Spectrophotometer and $\lambda_{\text{max}}=426 \text{ nm}$.

RESULTS AND DISCUSSION

The effects of Oxone® without catalyst, Na-Wy and the catalyst (CoAP) on tartrazine decolorization were monitored for the defined experimental conditions: 200 cm^3 of $C_{\text{dye}}=50 \text{ mg dm}^{-3}$ tartrazine solution, 40 mg of Oxone®, 0.1 g of catalyst, $50 \text{ }^\circ\text{C}$, 240 min (Figure 1).

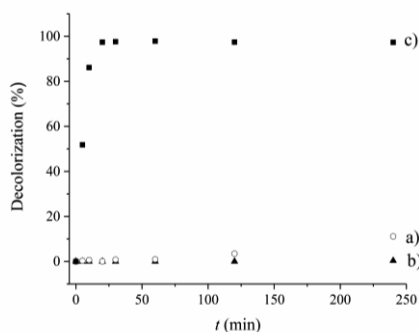


Figure 1. Decolorization of tartrazine a) Oxone® (without catalyst), b) Na-Wy, c) CoAP.

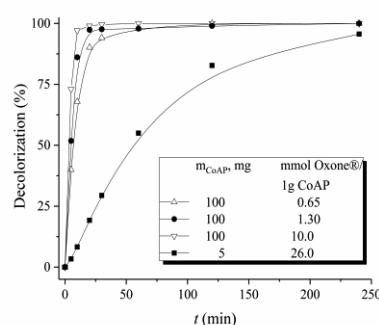


Figure 2. Decolorization of tartrazine with different ratios of Oxone® and catalyst.

The decolorization of tartrazine in the presence of the CoAP was almost complete after only 30 minutes (Figure 1) at the peak at 426 nm for the investigated conditions. On the other hand, in the reaction system without

catalyst, decolorization occurred, although very slow. After four hours the intensity of the initial color decreased for only 11 %. Reaction occurrence even without catalyst can be explained by the fact that Oxone® degraded tartrazine to some extent, even when there was no Co^{2+} to promote the creation of free radicals originating from Oxone®. It was probably due to selfdecomposition of Oxone® which led to formation of species that are able to oxidize although less efficient than radicals. In the case of the presence of Na-Wy in the reaction system decolorization was insignificant, indicating that Na-Wy did not show catalytic activity. Even more, Na-Wy, probably, adsorbed Oxone®, thus hindering the ability of Oxone® to decolorize tartrazine.

It can be observed (Figure 2) that, when the Oxone®/catalyst ratio was 10.0 mmol per 1g of catalyst, the reaction rate was the highest reaching 100% after 30 minutes. The drawback of these reaction conditions was that pH was 2.3 which was too low and resulted in undesirable level of Co^{2+} leaching into the reaction mixture. For the Oxone®/catalyst ratio of 26.0 mmol / 1g of catalyst, the rate of decolorization was the lowest because of lower amount of introduced catalyst (5 mg) in comparison with that in the other three investigated systems (100 mg). In this system pH was 3.5 and leaching was insignificant. Namely, it was below $20 \mu\text{g dm}^{-3}$.

CONCLUSION

Clay rich in montmorillonite was modified by means of Na-exchange process (Na-Wy) and subsequently pillared with aluminum. The obtained pillared clay was impregnated with Co^{2+} (CoAP). The CoAP was tested as catalyst in tartrazine solution decolorization. The role of catalyst was the activation of radical formation from potassium peroxydisulfate (Oxone®). The comparison of the effects of Oxone® without catalyst, Na-Wy and the catalyst (CoAP) on tartrazine decolorization revealed that after 30 minutes only in the CoAP+Oxone® system total decolorization occurred. The influence of the Oxone®/catalyst ratio on tartrazine solution decolorization was investigated and conditions for negligible impact of homogeneous catalysis were found. The cobalt impregnated pillared montmorillonite was found to be promising catalyst for dye decolorization.

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