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Hejda, S.
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Photocatalyzed degradation of dibutylphthalate with goethite and carboxylic acids

S. Hejda¹, O. Bajt², J. Krysa³, P. Kluson^{1,4}

¹ Department of Technical Sciences, Faculty of Environment, Jan Evangelista Purkyně University, Králova Výšina 3132, 400 96, Ústí nad Labem, Czech Republic

² National Institute of Biology, Marine Biology Station, Fornace 41, 6330 Piran, Slovenia

³ Department of Inorganic Technology, Institute of Chemical Technology, Prague, Technická 5, 166 28, Prague, Czech Republic

⁴ Institute of Chemical Process Fundamentals, Academy of Sciences of the Czech Republic, Rozvojová 135, 165 02 Prague 6, Czech Republic

Iron is one of the most abundant elements in the environment. Iron is present in different minerals, among them goethite, an iron oxohydroxide, is one of the most abundant. Different mechanisms for the photochemically enhanced reduction of Fe(III) have been proposed, but the most effective seems to be the photoreductive dissolution in the systems containing organic ligands, e.g. dicarboxylic acids. Dialkyl phthalates are used as plasticizers in different resins. They are considered to be toxic for different organisms; moreover, they are classified as important endocrine disrupting compounds.

The general experiment was performed as it follows. To the solution of dibutylphthalate (2.9×10^{-5} M) goethite was added (0.25 g L^{-1}). Suspension was homogenized in an ultrasonic bath, oxalic acid (1×10^{-3} M) was added to the homogenized suspension, and subsequently the reaction mixture was irradiated at 365 nm. The pH was kept constant using a pH-stat. Samples were taken in different time intervals, centrifuged and analyzed by HPLC.

During the experiments the effect of different parameters on the degradation rate was studied. Degradation was faster at lower pHs, up to pH 3, afterwards the degradation rate decreased with increased pH. The degradation at pH 7 after 3 h was almost negligible (< 10 %). The most efficient ligand was oxalic acid, with citric acid the degradation was much slower, and finally, the degradation with pyruvic acid as ligand accomplished less than 20 % after 3 h of irradiation. The degradation with salicylic, maleic and fumaric acids was almost negligible. The effect of the oxalic acid concentration was also observed. The degradation was faster at higher acid concentrations, but above 1×10^{-3} M concentration this effect became unimportant. Decrease in the goethite concentration led to the slightly slower degradation, while the opposite was observed with increased goethite concentration, but only up to 0.5 g L^{-1} . At higher concentrations the degradation rate decreased, probably due to the light scattering effect on goethite particles. Purging of the reaction mixture with air did not influence significantly the degradation of dibutylphthalate, while on the other hand, the presence of nitrogen resulted in decreased degradation rate. A continuous evolution of Fe(III) was observed, while the Fe(II) concentration was constantly low during degradation. This implies the formation of OH^\cdot after the photoreductive dissolution of Fe(II). Formation of OH^\cdot was confirmed by 2-propanol, an OH^\cdot scavenger.

In conclusion, the photocatalyzed degradation of dibutylphthalate under the presented conditions appeared as an efficient degradation process.