DEGRADATION OF PARACETAMOL IN AQUEOUS SOLUTION: COMPARISON OF DIFFERENT UV INDUCED ADVANCED OXIDATION PROCESSES

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Abstract — Environmental hazards are a consequence of the massive use of synthetic chemicals that are prevalent in nearly every aspect of modern life. The outburst of the so-called “emerging pollutants” (pharmaceuticals and pesticides among others) generates an additional problem due to the scarce available information on their impact on the environment or their interferences with the biological processes. Among them, paracetamol is one of the drugs the most widely found in hospital effluents, in the effluents of wastewater treatment plants, as well as in rivers. In this work, the feasibility of paracetamol degradation by different processes based on ultraviolet radiation is discussed: photolysis, photo-oxidation with hydrogen peroxide, photo-Fenton reaction, and heterogeneous photocatalysis with TiO$_2$ are evaluated. The best results are obtained with the photo-Fenton process, for which more than 99% of the pharmaceutical is degraded within 5 minutes. At best experimental conditions the mineralization was superior to 75% after one hour of reaction.

Keywords — acetaminophen, photolysis, UV/H$_2$O$_2$, photo-Fenton, UV/TiO$_2$.

I. INTRODUCTION

The rapid development of industry and society runs parallel with the increasing generation of wastewaters, containing highly toxic and poorly biodegradable compounds. This situation affects the efficiency of conventional biological treatments, the most commonly used processes in wastewater remediation. The so-called “emerging pollutants” (pharmaceuticals and pesticides among others) generate an additional problem due to the scarce available information on their impact in the environment or their interferences with biological processes (Fent et al., 2006). The presence of pharmaceutical compounds in water and wastewater has been repeatedly reported (Kümmeler, 2001; Ikhetat et al., 2006; Al Rifai et al., 2007; Gagné et al., 2006, Quesada Peñate et al., 2009a). Many of these active substances are persistent as they are transparent to conventional wastewater treatments and they are becoming ubiquitous in the environment (Carballa et al., 2004; Tauxe-Wuersch et al., 2005). Some of these compounds - in particular antibiotics, anti-tumor agents and analgesics - are indeed neither biodegradable, nor adsorbable on sewage sludge. Therefore, there has been an increasing concern about the impact of pharmaceutical compounds on public health and on the environment, not only because of their acute toxicity, but also their genotoxicity, as well as the development of pathogen resistance and endocrine disruption (Halling-Sørensen et al., 1998; Graham et al., 2011).

To drastically reduce the load of pharmaceutical compounds in discharged effluents, additional treatments have to be applied. Advanced oxidation processes (AOPs) have already proven to be effective for this purpose (Kolpin et al., 2002, Ikhetat et al., 2006; Al Rifai et al., 2007; McArdell et al., 2011). AOPs can be performed either as a tertiary treatment after biological (secondary) treatment, or as a pre-treatment stage in order to enhance the biodegradability of trace organic contaminants.

AOPs are characterized by a variety of radical reactions that involve combinations of chemical agents (e.g. O$_3$, H$_2$O$_2$), heterogeneous catalysts (transition metals and metal oxides) and auxiliary energy sources (e.g. UV-vis radiation, electronic current, and ultrasound). UV based AOPs include UV/H$_2$O$_2$, UV/O$_3$, UV/H$_2$O$_2$/O$_3$, UV/H$_2$O$_2$/Fe$^{3+}$ (or Fe$^{2+}$), and UV/TiO$_2$. The efficiency of the various AOPs depends both on the rate of generation of free radicals and the extent of contact between the radicals and the organic compounds (Gogate and Pandit, 2004). In the Fenton reaction, hydroxyl radicals (HO•) are generated by the catalytic decomposition of H$_2$O$_2$ using Fe$^{2+}$ in acidic medium (pH in the range 2–4) (Oliveros et al., 1997). Photo-Fenton oxidation is the photo-catalytically enhanced version of the Fenton process. In this process, UV light irradiation increases HO• formation rate, and also improves the regeneration of the ferrous catalyst by reduction of Fe$^{3+}$ and destruction of ferric complexes, thus allowing the