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Sonocatalytic Oxidation of EDTA using Noble Metal-Free Catalyst

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PURPOSE OF THE ABSTRACT

Among the various organic pollutants found in wastewaters, ethylenediaminetetraacetic acid (EDTA) is a very common one due to its use in many different fields like textile manufacturing, food or pharmaceutical industries. One way to treat this metal-mobilizing pollutant relies on Advanced Oxidation Process (AOP), which is the oxidation of organic compounds through the reaction with hydroxyl radicals or other strongly oxidative species [1]. Sonochemistry belongs to AOP family but its efficiency is usually limited to low concentrated organic compounds when used alone and required, for instance, the coupling with heterogeneous catalyst for more concentrated effluents [2-3]. Nevertheless, the catalysts used in that case are more often than not composed of noble metals which are expensive and scarce elements that should be replaced by earth-abundant materials [3]. This is from this perspective the current study is focused on the sonocatalytic oxidation of EDTA using a noble metal free catalyst based on Co_3O_4/TiO_2 system.

The EDTA degradation assays were carried out at high ultrasonic frequency of 345 kHz (Pac = 0.25 W/mL) under Ar/O_2 (20%) gas atmosphere at various temperatures (from 20 to 50 °C). These conditions are known to be in favor of the in situ generation of oxidative species like OH° and HOO° radicals leading to H_2O_2 formation with the reaction rate up to 9 µmol/min [4]. In all experiments, 2 g/L of Co_3O_4/TiO_2 catalysts were ultrasonically dispersed within 200 mL of 5 mM EDTA solution at low frequency and then submitted to 345 kHz ultrasound under mechanical stirring. The catalyst used in this study was prepared using a common impregnation method of cobalt nitrate on P25 TiO_2 prior room temperature drying and then calcination at 550°C with various amount of cobalt oxide ranging from 1.3 to 6.4 wt%. All the catalysts were characterized with HRTEM/EDX and XRD techniques before and after ultrasonic treatments. The concentration of EDTA and total amount of carbon in solution were monitored during all experiments using Fe-TPTZ dosimetry and TOC-meter analyses respectively. Obtained results indicate that Co_3O_4/TiO_2 catalyst is very stable under our conditions and shows better performance than Pt/TiO_2 catalyst. Moreover, 80% of the initial EDTA could be oxidized after 7 hours of ultrasonic treatment at 40°C while no significant EDTA degradation is observed in the presence of H_2O_2 without ultrasound. Finally, all these results allow us to suggest the EDTA sonocatalytic degradation mechanism depicted in Figure 1 involving the oxidization of EDTA on the Co^{3+} sites of Co_3O_4 and reactivation of the catalyst with in situ formed reactive species.

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FIGURES

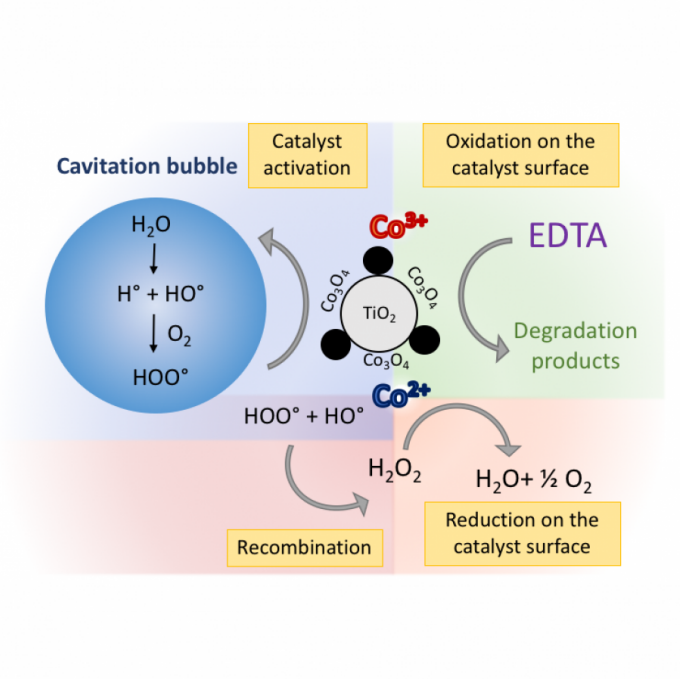

FIGURE 1

Figure 1

Mechanism of EDTA sonocatalytic degradation in the presence of Co3O4/TiO2 catalyst.

FIGURE 2

KEYWORDS

sonochemistry | EDTA | ultrasound | heterogeneous catalysis

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