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Catalytic wet oxidation of metal ion complexing agents by H₂O₂ using homogeneous and heterogeneous Fe (III) species

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A large amount of liquid radioactive wastes are formed during nuclear power plants operating. Radionuclides contained in the wastes are predominantly in the form of stable complexes with organic ligands, such as EDTA, citric and oxalic acids, which complicates the heavy metal separation. Concentrating and processing of liquid radioactive wastes can be greatly simplified after the removal of these organic compounds. Catalytic wet oxidation of complexing agents by H₂O₂ using Fe (III) species is a promising way for radioactive waste purification.

The presenting work considers a method of catalytic wet hydrogen peroxide oxidation (CWHPO) of EDTA using homogeneous and heterogeneous Fe(III) species. The solutions of mono- and polynuclear hydroxo complexes of Fe (III) have been obtained and studied as homogeneous catalytic systems. Several types of Fe (III) containing zeolites ZSM-5 with different particle morphology and texture, including nanozeolites and hierarchical zeolite, have been synthesized and tested as heterogeneous catalysts. The activity in CWHPO of different Fe (III) species was measured by barometric cell method and total organic content determination. In homogeneous 1 mM Fe (III) catalytic system the highest hydrogen peroxide decomposition rate was observed at pH = 2.5 when mononuclear Fe(III) hydroxo complexes were predominant. The reaction rate was found to be decreased at pH = 1.5 and 3.0 when hydrated and polynuclear hydroxo complexes were formed, respectively. At pH > 3 Fe (III) hydroxide precipitated and the reaction rate was negligibly low.

Unlike homogeneous system zeolitic one allows proceeding the reaction at a significantly wider pH range. Heterogeneity of the Fe (III) catalytic sites in the zeolite appeared to result in effective use of the hydrogen peroxide and a deep oxidation of a substrate. A comparative study of stability and activity of Fe (III) species supported onto the zeolite and amorphous SiO₂ has been made in oxidation of low and high molecular weight organic substances by H₂O₂. Catalytic site protection by zeolitic matrix plays a key role in the reactions and could be potentially explored for development of technology for purification of waste water from nuclear power plants.

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