Defence of a Doctoral Thesis

Metal-Chelate Complexes in Alkaline Solution:
On Recovery Techniques and Cellulose-based Hybrid Material Synthesis

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Abstract

For decades, aminopolycarboxylate chelating agents have been extensively used in various industrial applications. The ability of chelating agents to form stable metal-chelate complexes is the main reason for using them to manage metal ions within water-based industrial processes. Considerable quantities of industrial effluent containing chelating agents and heavy metals are produced and often discharged into the environment. The toxicity of heavy metals and the non-biodegradability of the chelating agents, as well as their accumulation in the environment, has become cause for concern. The main purpose of this thesis was to evaluate and develop processes for recovery of chelated metal complexes from aqueous solution. In this regard, the membrane electrolysis technique was evaluated for recovery of copper and aminopolycarboxylic chelating ligands such as ethylenediaminetetraacetic acid (EDTA), nitrilotriacetic acid (NTA), diethylenetriaminepentaacetic acid (DTPA), and a surface-active derivative of DTPA, 2-dodecyldiethylenetriaminepentaacetic acid (C_{12}-DTPA) from aqueous solution. By using this method, it was possible to simultaneously recover the chelating ligand for further reuse and collect the metals by electrodeposition, making the process more cost-effective and hindering the discharge of copper ions and chelating ligands as pollutants into the environment. In addition, the ion flotation technique with the chelating surfactant C_{12}-DTPA could be employed to separate metal ions, especially from their dilute solutions, and concentrate them in a foam phase. This is because C_{12}-DTPA has a purpose-built functionality; besides forming strong coordination complexes with metal ions, it is also surface-active and will readily adsorb at air-water interfaces. In this study, C_{12}-DTPA was effectively used in combination with foaming agents for the removal of toxic metal ions such as Cd^{2+}, Zn^{2+}, and Sr^{2+} from aqueous solution using ion flotation. From an economical perspective, this method could be combined with the membrane electrolysis technique to recover metal and regenerate chelating surfactant so that it can be reused.

The present work also shows the synthesis of metal and metal oxide(s) nanoparticles (NPs) in alkaline aqueous solution containing chelated metal ions, in order to fabricate metal NPs–cellulose hybrid materials. Cellulose is the most abundant renewable material, with good mechanical performance and chemical resistivity in a wide range of solvents, which makes it a promising material to support metal NPs. In this respect, we developed a rapid and inexpensive one-pot synthesis of spherical copper NPs in a cellulose matrix. The hybrid material displayed antibacterial properties for both the gram-negative and gram-positive bacteria. The synthesis was further developed by studying the influence of various chelating ligands and surfactants on the NPs' morphology and chemical composition. According to the results, DDAO, a zwitterionic surfactant, was found to mediate the formation of pure octahedral Cu_2O NPs. In addition, a hybrid material film composed of regenerated cellulose and synthesized Cu_2O nano-octahedrons was fabricated by spin-coating.

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