

Electrochemical Oxidation of Phenol Using Boron-Doped Diamond Electrodes

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Introduction

In recent years, there has been an increasing interest in the electrochemical properties of boron-doped diamond coated substrates. Silicon has been the predominate substrate used for diamond deposition. However, the conductivity of the silicon substrate is not sufficient for it to be used in higher current applications. This makes the use of metal wires or meshes as substrate materials highly desirable because of the increase conductivity and the possibility of increased surface areas. Recently, Glesener et al. have discussed the use of high surface area boron-doped diamond mesh for electrochemical applications (1).

The cleanup of liquids or slurries that contain >1% total organic carbon (TOC), defined as non-wastewater hazardous waste (2), offer particular difficulties for on-site remediation efforts. At the present time the preferred method of remediation for non-wastewater hazardous waste is by incineration.

The electrolytic destruction of organic wastes shows real promise for remediation of a wide variety of organic materials in aqueous waste streams (3). Use of electrochemical oxidation for organic waste decomposition is limited with the traditional electrode materials, platinum, ruthenium dioxide, lead dioxide and tin dioxide. These limitations arise from low reaction rates and efficiencies, corrosion of the electrodes and fouling and poisoning of the active electrode surfaces. The work reported here demonstrates that boron-doped diamond electrodes prepared by chemical vapor deposition (cvd) are not susceptible to these limitations and can be used to oxidize a wide variety of species, particularly phenol, completely to CO₂.

The oxidation of phenol was chosen as a test reaction because it is one of the most

difficult organic molecules to oxidize electrochemically (4). It is well known for its rapid fouling of the electrode surface due to formation of a blocking polymer layer produced by the polymerization of the phenoxy radicals generated in the initial stages of the reaction (5). This results in termination of the reaction in minutes.

Experimental

Electrodes were prepared by coating Ti mesh substrates (1 cm x 2 cm) with approximately 10 μm of boron doped diamond via microwave plasma enhanced chemical vapor deposition. A two step deposition process was utilized to achieve uniform nucleation and good film adhesion. The films were characterized with SEM and Raman to insure complete coverage of the Ti and to establish the quality of the cvd-carbon films.

The oxidation of phenol was studied with cyclic voltammetry in a solution of 0.1M H₂SO₄ containing 0.16 mM phenol in a beaker cell with a Pt counter electrode and a Pd-H reference electrode. Experiments were also run with a flow cell in which a liter of 10 mM phenol in 0.1M H₂SO₄ was circulated through the cell at 7ml/sec and the total organic carbon (TOC) was monitored as a function of time and cell current.

Results and Discussion

In our experiments phenol has been oxidized on boron-doped diamond electrodes without any reduction of the current arising from fouling or poisoning of the electrode even for concentrations of phenol which would put it in the category of non-wastewater hazardous waste. The total carbon in solution was reduced at high efficiency (80-90 %) from ~1% TOC to <0.1% TOC with no observable decrease in decomposition rate. This means that the reacted phenol was converted completely to CO₂. These results are extraordinary in that they are preliminary experiments on small surface area electrodes and the experiment has not been optimized in any way. In light of these results it would also be possible to treat a 10 % TOC solution as readily as a 1 % TOC solution.

Acknowledgements

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References

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