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Carbon-supported iridium catalysts in the catalytic wet air oxidation of carboxylic acids: kinetics and mechanistic interpretation

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Abstract

Carbon-supported iridium catalysts were prepared by different incipient wetness impregnation methods and by organometallic chemical vapor deposition. The catalysts were characterized by N_2 adsorption, TPD, SEM and H_2 chemisorption measurements. The results obtained indicate a clear dependency of the metal-phase dispersion on the pre-treatment of the carbon support and the impregnation method. Their activity for catalytic wet air oxidation of butyric and *iso*-butyric acid aqueous solutions was investigated in a stirred reactor at 473 K and 0.69 MPa of oxygen partial pressure. The conversions obtained after 2 h were 43 and 52%, with respect to each carboxylic acid, when the most active catalysts were used. The measured conversions and initial reaction rates correlate well with the exposed metal area. A rate equation was determined from measurements of the initial reaction rates at different oxygen partial pressures, temperatures and catalyst mass loads. The results were modeled considering a heterogeneously catalyzed free-radical mechanism.

Keywords: Catalytic wet air oxidation; Carbon-supported iridium catalysts; Incipient wetness impregnation; Organometallic chemical vapor deposition; Heterogeneous-catalyzed free-radical mechanism

1. Introduction

Wet air oxidation (WAO) is a useful process for reducing chemical oxygen demand (COD) of wastewaters, which can be used alone, or as pre-/post-treatment in combination with other processes, depending on the nature of the stream. In WAO, organic pollutants are usually mineralized to CO_2 and H_2O with an external oxidizing source (air or O_2) under conditions of high pressure and temperature [1–3].

Such conditions lead to very high investment costs, because reinforced materials are needed to support the strong pressures, and special metal alloys, which are much more expensive than commonly used stainless steel alloys, must be employed to avoid corrosion which is highly favored under these conditions. In spite of this, there has been a strong effort on the research and development of catalysts in order to bring down pressure and temperature to milder values—catalytic wet air oxidation (CWAO). Homogeneous catalysts such as copper [3–5] and iron salts [4] are

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