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Copper supported on porous activated carbon obtained by wetness impregnation: Effect of preparation conditions on the ozonation catalyst's characteristics

Un catalyseur au cuivre supporté par un charbon actif obtenu par imprégnation par voie humide : effet des conditions de préparation sur les caractéristiques du catalyseur d'ozonation

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ABSTRACT

An activated carbon-supported copper heterogeneous catalyst based on (Cu/AC) was developed using a wetness impregnation process. The effect of preparation conditions on the catalyst's characteristics was examined. This work focuses on two key parameters: impregnation rate and calcination conditions (temperature and time). Catalysts were characterized by means of nitrogen sorptiometry at 77 K, Boehm analysis and pH_{pzc} analysis. It was found that the catalyst properties and the functional surface groups were affected by the operating conditions. The highest measured surface area, i.e. 1040 m^2/g, was obtained for activated carbon (AC) impregnated with 12% of Cu loading after calcination at 550 °C for 2 h. The effect of adding copper on the surface of activated carbon on its adsorption capacity was also examined. The obtained results showed that after impregnation, the adsorption capacity of activated carbon was improved. Additionally, the performance of the Cu/AC catalyst on nitrobenzene ozonation was investigated. Our results show that the use of Cu/AC for heterogeneous catalytic ozonation enhanced significantly the degradation efficiency of nitrobenzene (NB) compared with simple ozonation and with ozonation catalyzed by AC without metal addition.

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RÉSUMÉ

Un catalyseur à base de cuivre supporté par un charbon actif a été développé en utilisant le processus d'imprégnation par voie humide. L'effet des différentes conditions de préparation sur la structure du catalyseur a été examiné. Dans ce travail, on s'intéresse à deux variables principales : le taux d'imprégnation et les conditions de calcination (température et temps). Différentes méthodes d'analyse ont été employées pour la caractérisation du catalyseur, à savoir l'isotherme BET, la méthode de Boehm et l'analyse

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du pH_{pzc}. Un optimum de la surface a été obtenu pour 12 % du cuivre supporté sur le charbon calciné à 550 °C pendant 2 h. L'effet du catalyseur sur la capacité d'adsorption du charbon actif a été examiné. Les résultats montrent que le greffage du cuivre à la surface du charbon actif a amélioré sa capacité d'adsorption vis-à-vis du nitrobenzène. Ce catalyseur a été par la suite testé pour l'ozonation du nitrobenzène. Les résultats montrent que la présence du catalyseur améliore considérablement la dégradation du nitrobenzène en comparaison de l'ozonation seule et de l'ozonation catalysée par le charbon actif.

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1. Introduction

The purification of water by ozone has been experimentally confirmed as the most appropriate way [1]. Ozone is known as a powerful oxidant of organic and inorganic micropollutants [2]. It acts either through a direct reaction of molecular ozone or through a radical-type reaction involving hydroxyl radical (OH°) generation induced by ozone decomposition in water [3]. However, ozone reacts slowly with some organic compounds such as nitro-aromatic compounds. For this reason, many works focused on developing catalysts for ozonation in order to enhance ozone's action and to achieve higher oxidation efficiencies. The combination of ozone with homogeneous or heterogeneous catalysts, with or without metallic compounds, has been investigated [4]. Adding a catalyst can improve the oxidation rate and the ozone concentration required to achieve the same degradation level [5]. Metal ions in acidic solution and metal oxides are usually used as catalysts for ozonation [6]. Using metal salts dissolved in solutions or solid powder metal oxides can cause secondary polluting problems because catalysts have to be removed after the oxidation reaction of the organic compounds. Those particles are unstable and will aggregate together to form large particles. These problems can be overcome and the performance of such catalysts can be improved by highly distributing the catalyst components over a porous support [5–7]. The advantages of using catalysts supported on porous materials are as follows: the active agent could be kept in a dispersed and stable state, the organic compound to be oxidized can be adsorbed onto the porous support at first to enhance its surface concentration, which is favored by the catalytic reaction as it accelerates oxidation during the reaction. The oxidation reaction occurs at the support surface so that the reaction activation energy can be reduced; so the residence time for reaction completion can be shortened. As a consequence, the investment and operation costs can be reduced [6].

The use of carbonaceous materials as catalytic supports continues to increase because of the high versatility of this material [8]. The most important carbon support material is activated carbon (AC), followed by carbon black and graphite materials [6]. Based on its porous structure and surface functional groups, activated carbon exhibits the most favorable physical and chemical characteristics [9] for the preparation of supported catalysts [8,10–13]. The active metals will be dispersed into the small pores; oxidized intermediate products could achieve total oxidation, giving as final products carbon dioxide and water [6]. In the view of these characteristics, activated carbons are used as supports for a great number of active phases, including noble metals [7,8,14,15]. In catalytic processes, metal dispersion in a final catalyst plays a key role as it enhances its efficiency [16]. The interaction of the support surface both with the metal precursor and with the solvent during the impregnation step greatly influences metal uptake and dispersion into the catalyst [8]. It is necessary to maximize the dispersion of the metal on the surface of the support. The porous structure controls the availability of surface active sites and thereby the degree of catalyst dispersion. Pore size distribution plays a crucial role in catalyst preparation. The access of metals can be also improved by surface groups due to the decrease of the hydrophobicity of the carbon [16]. There are only a few studies, which were interested in investigating the effect of preparation conditions on the dispersion of metals and the structure of the final catalyst [6,8].

The use of a supported catalyst is common in the oxidation of a great number of organic compounds [17–22], such as nitrobenzene [23,24]. This molecule is considered to be highly toxic; unfortunately, it is resistant to oxidation by biological treatment processes due to its carcinogenesis and mutagenesis [24]. It is also considered as a hard biodegradable compound and inhibitor for activated sludge [25]. This important class of industrial chemicals is widely used in the synthesis of many products, including dyes, polymers, pesticides, and explosives [26,27]. Unfortunately, the widespread application of nitro-aromatic compounds and their improper disposal has resulted in their release into the environment [28]. The nitro group, which provides chemical and functional diversity in these molecules, also contributes to the recalcitrance of these compounds to biodegradation [27]. There is abundant literature [23,29] dealing with the oxidation of nitrobenzene using Cu-based catalysts. The results indicate that supported catalysts show significant activities for the catalytic oxidation of nitrobenzene.

The aim of this work is to investigate the effects of preparation conditions, metal precursor nature and texture of the support on the properties of the final catalyst, its adsorption capacity, and its catalytic activity in the ozonation of nitrobenzene.

2. Experimental

2.1. Materials and reagents

A commercial granular activated carbon (AC), type Chemviron CAL, produced from selected-grade bituminous coal by a highly developed and strictly controlled

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