Improving Ozone Water Purification Efficiency with Green Catalysts

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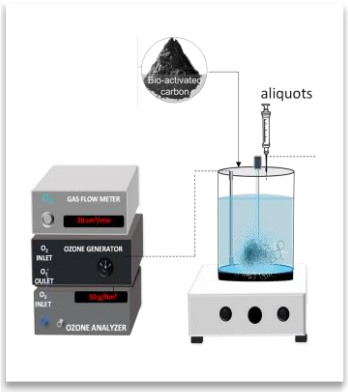
C. Graça[1,2], O. Soares[1,2], (1) Laboratory of Separation and Reaction Engineering - Laboratory of Catalysis and Materials (LSRE-LCM), catiaalgraca@fe.up.pt. (2) ALiCE – Associate Laboratory in Chemical Engineering, Faculty of Engineering, University of Porto, Rua Dr. Roberto Frias, 4200-465 Porto, Portugal



Cork residues were transformed into activated carbons through physical activation with carbon dioxide (CO_2). The main goal was to potentialize the application of this waste-derived activated carbon as catalysts in ozonation reactions for water remediation purposes. The key parameters of the physical activation, namely the activation temperature, dwell time and $\%CO_2$, were optimized according to a Box-Behnken experimental design, considering the catalytic activity of the produced materials in the ozonation of oxalic acid (OXL) as the response. Optimal conditions were found for the thermal treatment of cork residues that enabled achieving 92% of OXL removal, almost 5 times higher than that observed with single ozonation, namely 500 °C of activation temperature, 2 h of dwell time and 70% of CO_2. This study intends to offer a sustainable approach to waste management while bringing an alternative solution for water treatment challenges.

Introduction

Ozone (O_3) is a powerful oxidant already employed worldwide for water treatment and disinfection, given its recognized effectiveness against bacteria, viruses, and organic pollutants. However, O_3 presents limited activity towards some small-chain degradation products, namely deactivated aromatic or electron-poor aliphatic pollutants, under the conditions employed in water treatment, leading to their accumulation in the environment. This is the case of oxalic acid (OXL), a typical ozonation by-product that accumulates due to its refractory character towards O_3. A possible strategy to remove this kind of refractory by-products would be the combination of O_3 with catalysts capable of transforming O_3 into more reactive species. Our group has been exploring green catalytic alternatives to perform this process, such as metal-free alternatives (*e.g.* biomass-derived activated carbon).These are considered more economical and environmentally friendly options than other catalysts used in this process, since they can be made from inexpensive or/and 100 % natural sources. In this work was explored the potential of cork waste-derived activated carbon as catalyst in the ozonation of OXL.

Material and Methods

Cork wastes (CK) were used as precursor for the production of activated carbons (Acs). These were provided by a cork producer company located in Portugal. These residues were dried in an oven at 110 °C for 24 h before being submitted to the activation process.

Activated Carbons were prepared by one step physical activation that consisted in subjecting the cork residue to pyrolysis under an inert atmosphere (N_2) containing different proportions of CO_2. The treatment time, temperature (°C) and $\%CO_2$ were varied according a design of experiments, as a strategy to find the optimum conditions for catalyts fabrication that resulted in higher catalytic activity for OXL removal through ozonation. Treatment times were varied between 1, 2 or 3 h, for three different dwell temperatures (500, 600, 800 °C) and three distinct $\%CO_2$ (30, 50 and 70% v/v), which gives a total of 12 different materials according to a Box-Benkhen design approach. Experiments with materials prepared using the conditions of the central levels (i.e. 2 h at 600 °C with 50% CO_2) were repeated 5 times for statistical validation. Obtained activated carbons were analyzed for their surface area by the Brunauer, Emmett and Teller method (S_{BET}). To perform the reactions, 50 mg of catalyst was added to a solution containing 20 mg L^{-1} of OXL and the reaction started when a stream of O_3 was bubbled into this suspension. The mixture was continuously stirred during 180 min and aliquots were taken at determined times to determine OXL concentration. Experiments in the absence of catalyst (single ozonation) or O_3 (adsorption) were performed for control purposes. An illustration of the experimental apparatus is depicted in the graphical abstract.

Results and Discussion

Most of the materials prepared enabled accelerating OXL ozonation. A straight correlation was observed between the materials' surface area (S_{BET}, $m^2.g^{-1}$), activation temperature and OXL adsorption capacity, as expected. However, the contrary was observed for the results of catalytic ozonation. In this process, the materials' activation temperature presents a negative effect, i.e. increasing the temperature of the thermal treatment decreases the catalytic performance of the activated carbon. Previous studies indicate that when the activation temperature is too high, the biomass-derived activated carbon loses microporous structures and also becomes more acid, which is known to negatively impact the catalytic performance in ozonation reactions [1, 2]. Figure 1 depicts the response surface of OXL% removal in 180 min of ozonation catalized by the produced

activated carbons from cork waste. The response surface has a convex shape, with a minimum at the central conditions (600 °C for 2 h under 50% CO_2). The best catalytic performance (92% OXL removal) was observed with the cork residue treated at 500 °C, for 2 h and under 70% of CO_2. Further investigation is now required to explain these results, such as textural and surface chemistry analyses of the prepared activated carbons.

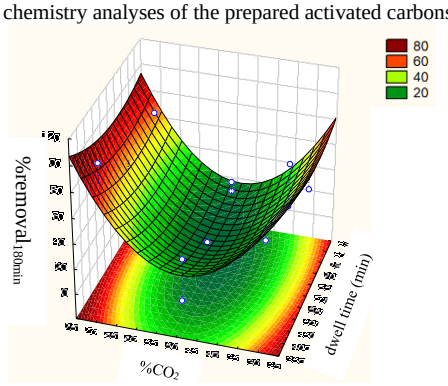


Figure 1. Response surface of the %OXL removal in 180 min reaction, varying the activated carbon synthesis parameters (%CO_2 and dwell time). Thermal treatment temperature = 600 °C.

Conclusions

This study underscores the significant catalytic potential of activated carbons derived from cork waste in ozone-mediated water treatment. The enhanced performance of residues thermally treated at lower temperatures highlights the promising role of these sustainable precursors for efficient and eco-friendly water treatment processes. Such observations encourage further research to explore and optimize the catalytic capabilities of these biomass-derived activated carbons.

Acknowledgments

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