UNIVERSITI TEKNOLOGI MARA

DEGRADATION OF ORANGE II DYE USING PEROVSKITE COMPOUNDS WITH MOLYBDENUM ION AS ACTIVE PHASE

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ABSTRACT

Heterogeneous chemical oxidation is a promising technique and effective treatment in degrading dye-contaminated wastewater which generated highly oxidizing radicals such as hydroxyl (˙OH) radicals. Perovskite is a potential decent ceramic metal oxides catalyst with the general formula of $AB_xB'_yO_3$ has been applied as a heterogeneous catalyst for dye-contaminated wastewater degradation under ambient conditions with and without the aid of oxidants. Substitution of B-site cation plays a crucial role in modulating the overall catalytic performance of the perovskite rather than A-site cation. Despite its high catalytic performance, these catalysts suffered severe leaching problem during heterogeneous catalysis. Therefore, substitution of alternative B-site metal cations in perovskite to minimize the leaching problem while maintaining adequate catalytic performance is critical. However, the substitution of molybdenum as an active phase for B-site cation within the perovskite structure is still scanty. Hence, this research aims to synthesize and characterize substituted molybdenum derived perovskite catalysts and investigate the resultant catalyst's stability at the optimum oxidative degradation conditions. A series of substituted molybdenum derived perovskite catalysts ($CaMo_xCu_yO_3$) were synthesized using the EDTA-citric acid complexation method by varying the precursor loading molar ratio (Mo:Cu = x:y). The catalytic activity of resultant catalysts was evaluated in oxidative degradation using acid orange II (OII) dye solution as a model pollutant. The perovskite catalysts were characterized using field emission scanning electron microscopy, nitrogen sorption, X-ray diffraction, chemisorption analysis. Response surface methodology (RSM) facilitated the operational conditions' optimization for OII oxidative degradation at the optimum composition of $CaMo_xCu_yO_3$. Then, the catalyst's stability was subsequently tested at optimum operational reaction conditions. The optimum perovskite composition was found at $CaMoCu_{0.5}O_3$, which exhibited 46.43% OII removal without the aid of an oxidant. Interestingly, the removal of OII solution increased two times higher up to 98.43% in the presence of oxidant within 30 minutes reaction compared to others composition loading ratio. From RSM analysis, optimum operational reaction conditions were proposed at catalyst dosage of 0.99 g L^{-1}, 20.80 mg L^{-1} initial concentration of OII solution, 5.42 pH of the solution and 0.87 g L^{-1} concentration of H_2O_2 at 15 min of reaction time. The experimental validation of OII removal was found to be approximately 91.19% at the proposed optimal conditions. The experimental OII removal values were aligned with the predicted value derived from the proposed quadratic model with a slight difference of 3.98%. Uniquely, the $CaMoCu_{0.5}O_3$ perovskite exhibited a slight decrease in the overall catalytic performance by 26% in the first three cycles, which stabilized afterwards until the fifth cycle of reaction with minimal leached metal ions' concentration within the allowable regulatory limits. These results provide new insights into the unique role of substituted molybdenum as an active phase in enhancing the overall catalytic performances for oxidative degradation of dye-contaminated wastewater. In summary, findings of this work support the Malaysia's Twelfth Malaysia Plan and United Nations' goal in improving access to safe and clean water.

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