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## **ACCEPTED MANUSCRIPT**

Screening and optimization of highly effective ultrasound-assisted simultaneous adsorption of cationic dyes onto Mn-doped  $Fe_3O_4$ -nanoparticle-loaded activated carbon

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#### **Abstract**

The ultrasound-assisted simultaneous adsorption of brilliant green (BG) and malachite green (MG) onto Mndoped Fe<sub>3</sub>O<sub>4</sub> nanoparticle-loaded activated carbon (Mn-Fe<sub>3</sub>O<sub>4</sub>-NP-AC) as a novel adsorbent was investigated and analyzed using first derivative spectrophotometry. The adsorbent was characterized using FT-IR, FE-SEM, EDX and XRD. Plackett-Burman design was applied to reduce the total number of experiments and to optimize the ultrasound-assisted simultaneous adsorption procedure, where pH, adsorbent mass and sonication time (among six tested variables) were identified as the most significant factors. The effects of significant variables were further evaluated by a central composite design under response surface methodology. The significance of independent variables and their interactions was investigated by means of the analysis of variance (ANOVA) within 95 % confidence level together with Pareto chart. Using this statistical tool, the optimized ultrasoundassisted simultaneous removal of basic dyes was obtained at 7.0, 0.02 g, 3 min for pH, adsorbent mass, and ultrasonication time, respectively. The maximum values of BG and MG uptake under these experimental conditions were found to be 99.50 and 99.00 %, respectively. The adsorption process was found to be followed by the Langmuir isotherm and pseudo-second order model using equilibrium and kinetic studies, respectively. According to Langmuir isotherm model, the maximum adsorption capacities of the adsorbent were obtained to be 101.215 and 87.566 mg g<sup>-1</sup> for MG and BG, respectively. The value of apparent energy of adsorption obtained from non-linear Dubinin-Radushkevich model (4.348 and 4.337 kJ mol<sup>-1</sup> for MG and BG, respectively) suggested the physical adsorption of the dyes. The studies on the well regenerability of the adsorbent in addition to its high adsorption capacity make it promising for such adsorption applications.

**Keywords:** Basic dyes removal; Derivative spectrophotometry; Experimental design; Isotherms and kinetic studies; Mn-Fe<sub>3</sub>O<sub>4</sub>-NPs-AC; ultrasound-assisted simultaneous adsorption.

#### 1. Introduction

Wastewater generated from textile, leather, paper and plastic industries contains toxic dyes, which can make serious damages on water resources and can thus results in severe economic and environmental problems [1, 2]. Brilliant green (BG) and malachite green (MG) as cationic dyes are highly water-soluble and used as coloring agent in textile, silk, paper, dermatological agent, biological stain, veterinary medicine as well as for painting

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