

**UNIVERSITI TEKNOLOGI MARA**

**MAGNETIC BENTONITE  
COMPOSITE FOR  
DECOLORIZATION OF  
METHYLENE BLUE AND METHYL  
ORANGE DYE IN AQUEOUS  
SOLUTION**

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## ABSTRACT

Magnetic bentonite composite is one of the recent engineered clay materials currently developed for the treatment of dye-contaminated water. However, its efficiency against different type of dyes, structural transformation and stabilities, and practicality in treatment of real effluent are relatively unknown. This study aims to prepare the magnetic bentonite composite (MBC) using co-precipitation technique by combining bentonite and iron oxide (IO) via *in situ* approach. The composite's structure, morphological and magnetic properties were characterized using spectroscopic, macroscopic and various analytical techniques like Fourier transformed infrared (FTIR) spectroscopy, X-ray Powder Diffraction (XRD), X-ray Fluorescence (XRF), Vibrating Sample Magnetometer (VSM), Scanning Electron Microscopy coupled with Energy Dispersive X-ray spectrometry (SEM-EDX), Transmission Electron Microscopy (TEM), surface area analysis using N<sub>2</sub> adsorption-desorption isotherm and pH<sub>zpc</sub> analysis. The removal efficiencies of MBC against the cationic (Methylene Blue, MB) was investigated against experimental parameters such as pH, initial dye concentration, and contact time followed by comparison with an anionic (Methyl orange, MO) azo dye. The desorption efficiencies were investigated using different eluents such as NaCl, NaOH, HCl, ethanol, and deionized water for reusability studies. The stability and structural transformation of spent and reused MBC were elucidated via FTIR, XRD, SEM, TEM, and VSM analysis. The practicality of MBC towards removing MB in more realistic water conditions was further assessed by adsorption experiment in the artificial textile effluent. The characterization results showed that the magnetic composite exhibited a saturation magnetization of 10.6 emu/g with high degree of crystallinity as supported by the XRD analysis. The composite shows better performance (100% dye removal) towards MB removal as compared to the MO dye. The equilibrium adsorption profile has a good agreement with the Langmuir isotherm model ( $R^2$ : 0.9982) and followed the pseudo-second order kinetic model ( $R^2$ : 0.999). The proposed mechanism of MB adsorption on MBC were via the hydrogen bonding,  $\pi$ - $\pi$  stacking and electrostatic attraction. Desorption of spent MBC (SMBC) using 0.1 M NaCl recorded the highest desorption efficiency (64%). SMBC demonstrates a high structural stability and minor morphological changes as evidenced by the characterization data. Three (3) adsorption-desorption cycles were successfully carried out during reusability studies. However, the magnetic strength and removal efficiency of the reused MBC (RMBC) declined to 5.5 emu/g and 50% after the final cycle. The MBC shows a 100% removal of MB in artificial textile effluent with improved water quality parameters. This composite has a great potential as a green and efficient adsorbent for water remediation especially for treating dye contaminated water. The research findings on structural transformation of magnetic clay contributes to the knowledge advancement of the environmental sustainability of engineered clay adsorbent.

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# CHAPTER ONE

## INTRODUCTION

### 1.1 Research Background

An economical and reliable wastewater treatment for industrial effluent is one of the major environmental challenges faced by developing countries in keeping abreast with emerging socio-economic growth. Various naturally derived adsorbents such as activated carbon, fly ash, and bamboo charcoal have been studied for removing pollutants from wastewater (Ray et al., 2022). However, the high operational cost, inefficient sorption capacity, and poor regeneration feasibility of the aforementioned adsorbents limits their potential application on industrial scale (Saeed et al., 2020). On the other hand, clay minerals resources (e.g., bentonite, kaolinite, and montmorillonite) received great interest in many environmental studies because of its low cost, high mechanical and thermal stability, and huge abundance (Kadeche et al., 2020). Bentonite clay mineral is a natural aluminosilicate typically found in many countries like France, China, and Indonesia (Maryam, 2017). Smectite (more widely known as montmorillonite) makes up at least 50% of the composition of the bentonite. According to Çiftçi, (2022) and Obiageli, (2017), bentonite is one of the favourable adsorbent in water remediation; especially for organic contaminants (Santos et al., 2020) due to its large specific surface area, as well as high availability with good chemical and mechanical stability (Lou et al., 2017). Bentonite also has high cation exchange properties as compared to kaolinite and sepiolite. The surface of clay minerals is naturally negatively charged which provides excellent characteristics for decolorization of cationic contaminants like methylene blue (MB) azo dyes (Sarkar et al., 2018). Despite those advantages, spent bentonite adsorbent is difficult to recover from the suspension media due to its fine particles' character, leading to reusability and sludge build-up issue.

Meanwhile, inorganic nanoparticle like iron oxide (e.g., magnetite  $\text{Fe}_3\text{O}_4$ ) prepared via the co-precipitation method is also used for the degradation of organic contaminants like anilines, phenols, nitrophenol and dye (Sahoo et al., 2019). The magnetic susceptibility feature of this iron oxide promotes its widespread applications especially for the separation and recoveries of spent adsorbents (Ain et al., 2020). The