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Removal of Microplastics using Ground Granulated Blast Furnace Slag

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Abstract **-** In this study, microplastic removal using ground granulated blast furnace slag was investigated. Blast furnace slag is a waste material that requires sustainable disposal or reuse alternatives. Microplastics of polyethylene have been the focus of this study. Batch adsorption experiments were conducted to evaluate the ability of ground-granulated blast furnace slag to adsorb and remove microplastics from water. Primary microplastic particles were procured in different sizes and colors for this study to ensure quality control. Experiments were conducted with 2, 4, 6, 8, and 10 g/L of ground granulated blast furnace slags were used in the study. All the experiments were conducted with 500 mg/L of microplastic concentrations. The experimental setups were operated on the shaker table and waited until they reached equilibrium. Standard analytical methods were used to test microplastic concentrations. Even though the experimental results indicated effective microplastic removal using ground granulated blast furnace slag, Fourier Transform Infrared Spectrometer couldn't identify and recognize the adsorption process with clarity. The experimental results indicated that an increase in the blast furnace slag decreased the microplastics concentrations. It was revealed that the increase in the microplastic size reduced the adsorptive capacity of the ground granulated blast furnace slag. The results also indicated that the freundlich isotherm model was better suited to represent the adsorption process than the Langmuir isotherm model. The study concluded that ground granulated blast furnace slag has the potential for microplastic removal from water. However, further investigation will be needed.

Keywords**:** Microplastics, Water Pollutant, Adsorption, Water Treatment

1. Introduction

A significant number of plastic items are discarded after use daily. A total of 320 million tonnes of plastic waste are discarded annually, 10% of which eventually reaches and persists in aquatic environments [1]. Due to the influence of physical chemical, and biological factors in the environment, plastic can degrade into five various sizes and forms, including nanoplastics (≤0.1 µm), microplastics (<5 mm), mesoplastics (0.5–5 cm), macroplastics (5–50 cm), and megaplastics (>50 cm) [2].

Microplastics (MPs), which have become increasingly problematic in recent years, are generally classified as primary and secondary [3]. Primary MPs are directly manufactured plastic particles, serving various purposes in products like cosmetics, films, textiles, and more. Secondary MPs result from the breakdown of larger plastic items. The widespread use of plastic is due to its low cost, versatility, and durability, making it prevalent in many industries and daily life.

Due to plastic's popularity, its production rate has increased significantly compared to most other synthetic materials. However, MP pollution poses severe threats, causing harm to fish and ecosystems, with far-reaching consequences that are challenging to quantify. MPs can impact the human body by stimulating the release of endocrine disruptors. Additionally, MPs can carry other toxic chemicals such as heavy metals and organic pollutants during adsorption which can adversely affect the human body. Given their ability for adsorption, microplastics have gained interest as a solution for the removal of other contaminants in water.

One of the contaminants that can be adsorbed by MPs is heavy metals. Around the world, heavy metals are widely considered the most significant environmental pollutant due to their extensive pollution sources [4](Rehman et al., 2017). Heavy metals can contaminate the environment through various human activities, such as mining, smelting, electroplating, and the production of paint, dyes, and other industrial products. Additionally, solid waste and domestic sewage can also contribute to heavy metal contamination.

Among the most common heavy metals are Cd^{2+}, Co^{2+}, Cu^{2+}, Ni^{2+}, Pb^{2+}, Zn^{2+}, and Hg^{2+}. These metals are characterized by low degradability and high toxicity [5]. In a research investigation that explored the adsorption properties of heavy metals onto microplastic, both in controlled laboratory experiments and field tests, the findings revealed that specific categories of

MPs exhibited varying levels of absorptive capacity for distinct heavy metals, and these variations were correlated with factors such as ion concentration, adsorption duration, and particle size [6].

In another study, researchers examined how three types of MPs with similar particle sizes, namely Polypropylene (PP), polystyrene (PS), and polyvinyl chloride (PVC) interacted with trace heavy metals like Pb^{2+}, Cu^{2+}, Cr^{6+}, and Cd^{2+}. They investigated the adsorption mechanisms under temperature and salinity conditions. The results revealed that the primary factor influencing metal adsorption onto MPs was particle diffusion. The isotherm model suggested that the adsorption of Pb^{2+}, Cu^{2+}, $Cr^{6+,}$ and Cd^{2+} onto MPs followed a physical monolayer adsorption process. Additionally, it was observed that higher temperatures and lower salinity levels enhanced the affinity between MPs and heavy metals through adsorption [7]. The affinity of MPs for heavy metals is a concerning topic, as MPs in the environment can transport these heavy metals and release them into water bodies or food chains if ingested by animals. Controlling the release of MPs into the environment is therefore necessary. This is because conventional wastewater treatment plants are not fully capable of removing MPs, highlighting the need for the development of efficient, simple, and low-cost strategies for MP removal from wastewater to prevent their entry into water bodies.

Since MPs have a high affinity for metals, the effect of metals on the adsorption of MPs is a topic of interest in the literature. Wang et al. (2021) investigated the effectiveness of magnetic biochar adsorbents that have been modified with Mg and Zn to eliminate microplastics [8]. When tested with 1 μm polystyrene (PS) microspheres in a water-based solution at a concentration of 100 mg/mL the removal rates, for biochar (MBC), Mg-modified magnetic biochar (Mg MBC), and Zn-modified magnetic biochar (Zn MBC) were 94.81%, 98.75%, and 99.46% respectively. Studies on similar adsorbents, and the adsorption of MPs in general, remain limited, even though adsorption is an effective and low-cost treatment option. In a study conducted by Sundbaek et al. (2018), they examined how fluorescent MP particles adhere to the surface of a microalgae called Fucus vesiculosus also known as seaweed [9]. The size of the microplastics was 20 μm while the plant cells of the seaweed had narrow microchannels that limited the movement of polystyrene microplastics into its tissues. The findings showed an absorption of MPs (94.5%) primarily near the cut surfaces of the seaweed.

Moreover, in another study that investigated the ability of oat protein sponges to capture MPs, the results showed that hydrophobic interactions and diffusion within the sponge played a crucial role in the adsorption process. The sponge had a structure that was well connected making up 83% of its composition. Consequently, it displayed a capacity to rapidly absorb MPs with as much, as 38% being captured by the sponge in just 10 seconds [8].

Various studies in the field of adsorption explored various materials as potential adsorbents. A recent and innovative research avenue focuses on utilizing solid waste generated during iron and steelmaking processes to extract contaminants from polluted water. One such waste material is blast furnace slag, which contains significant amounts of metal oxides and possesses notable porosity, specific surface area, and strong adsorption capabilities [10]. Leveraging these adsorption properties ferrous slag has been applied as a cost alternative to activated carbon in water and wastewater treatment. During the water purification process, ferrous slag employs not only physical adsorption but also various chemical processes such, as reduction, precipitation, coordination exchange, and ion exchange. These mechanisms work together synergistically to eliminate types of impurities.

Abdelbasir and Abdel Khalek (2022) investigated the use of blast furnace slag (BFS) as an affordable adsorbent to remove heavy metal ions particularly Co^{2+} and Pb^{2+} from water solutions [11]. Their findings revealed that when exposed to pH 6 conditions for 60 minutes BFS demonstrated excellent adsorption capabilities with Co^{2+} reaching up to 43.8 mg g−1 and Pb^{2+} reaching up to 30.2 mg g−1. The adsorption kinetics for Co^{2+} followed the Avrami model while the Freundlich model proved suitable for describing the adsorption isotherms of Pb^{2+}.

Despite the studies that have been carried out on blast furnace slags' ability to absorb various substances there has been limited research focused on its potential to adsorb MPs. Specifically, there is a lack of investigations into using this material for removing MPs. Given the metal content in Ground Granulated Blast Furnace Slag (GGBS) and its well-known adsorption properties, it offers an exciting avenue for further exploration, especially considering the strong affinity between MPs and heavy metals. On average, the chemical composition of GGBS consists of 43.8% active calcium oxide (CaO), 37.7% active silicon oxide (SiO_2), and 10.2% aluminum oxide (Al_2O_3). Iron oxide (Fe_2O_3) and other compounds comprise the remaining proportions of its composition [12]. The objective of this study is to bridge this knowledge gap by conducting batch adsorption tests to evaluate the capacity of GGBS in adsorbing MP of PE of three different sizes.

2. Materials and Methods

2.1 Materials Characterization

Microplastics were purchased from Cospheric Inc. in three different average particle sizes, where each had a different particle color: 125-150 micrometers (blue), 250-300 micrometers (red), and 425-500 micrometers (green). The microplastics particles are mostly round and are light in weight, hence they tend to float in water. GGBS was obtained from a local supplier as a byproduct of the steel manufacturing industry. The GGBS is whiteish-grey in color, and its composition was characterized through a Fourier Transform Infrared Spectroscopy (FTIR) analysis. The interaction between GGBS and microplastics in aquatic environments was investigated through batch adsorption experiments.

2.2 Experimental Setup

The adsorption experiment was undertaken where the adsorbent, GGBS, was used to capture and remove the adsorbate, MPs. The adsorption experiment was performed for the three different sizes of microplastics using five varying doses of GGBS: 2, 4, 6, 8, and 10 g/L. A constant concentration of microplastics (500 mg/L) was added to each test to create synthesized contaminated solutions. To conduct a bench scale test representing the adsorption experiment, 200 mL of water was used and the GGBS doses were converted correspondingly to 0.4, 0.8, 1.2, 1.6, and 2 g per 200 mL of distilled water. Before conducting the test, several trial-and-error experiments were undertaken to establish an approximate equilibrium state for the adsorption of MPs with GGBS. Accordingly, an experimental methodology was defined.

2.3 Experimental Procedure

The samples were mixed 6 g/L of GGBS and shaken at 350 rpm to determine the equilibrium time. From 0 to 120 minutes, at intervals of 20 minutes, a representative sample was taken out and filtered to check for the MPs retained in the solution. The equilibrium time was subsequently determined as the time when the concentration of MPs in the treated solution remained constant.

After the preparation of the synthesized solutions and the addition of the GGBS, the samples were placed on a lab shaker table to ensure sufficient mixing of the materials. The shaker table was operated at 350 rpm for the equilibrium time. After mixing, the pH of each solution was measured using a pH meter and the solutions were divided into 3 samples of each solution configuration. All samples were then placed in a centrifuge for 10 minutes to separate the GGBS from the solution to sample the equilibrium concentration. The centrifuge was set to 2500 rpm and the temperature was maintained at 22°C. The samples were then filtered using the lab filtration vacuum pump. To account for the MPs removed during centrifuging, a blank solution containing only MPs was placed in the centrifuge, and the solution was filtered to check the MP concentration.

2.4 Analytical methods

Adsorption in this experiment was evaluated based on the mass of the MPs remaining in each sample, which, in return, is related to the mass of MPs adsorbed and removed from the solution. Hence, the filter papers were weighed before and after filtration, and the mass of MPs that were not removed by adsorption was obtained.

2.5 Adsorption isotherms

The percent removal of MPs is estimated using the eqn (1) [11]

$$\text{Removal Efficiency } \% = \frac{C_i - C_f}{C_i} \text{ x } 100 \qquad (1)$$

Where C_i and C_f are the initial and final MP concentrations (grams per liter)

The weight of adsorbed MPs per unit weight of GGBS, q, (mg/g), is determined by the below equation:

$$\text{Adsorption Capacity, q, (mg/g)} = \frac{(C_i - C_f) \text{ x } V}{W} \qquad (2)$$

V is the solution volume (L)

W is the GGBS dose (grams)

To determine the adsorption capacity of the GGBS. The following isotherms were used in this study.

Langmuir Isotherm

$$\frac{C_e}{q_e} = \frac{1}{Q_m K_L} + \frac{C_e}{Q_m} \qquad (3)$$

Where Q_M is the monolayer saturated adsorption capacity (mg/g) and K_L refers to the Langmuir adsorption equilibrium constant (L/mg)

Freundlich Isotherm

$$q_e = K_f C_{e^{1/n}} \tag{4}$$

Where Kf is Freundlich capacity factor $(mg/L)^{-1/n}$

1/n is the Freundlich intensity parameter

3. Results and Discussion

3.1. Characterization of GGBS

The FTIR spectrum of the GGBS is shown below. The peak at 2872 cm^{-1} can be attributed to the presence of calcium carbonate while that at 875 cm^{-1} can be attributed to the vibration of the aluminate bond. The check against the available library also confirmed the presence of magnesium oxide. The analysis was done for the GGBS precipitate post-adsorption, but as the sample was not separated effectively from the solution, the spectrum was that of water, and no peaks related to GGBS were noted.

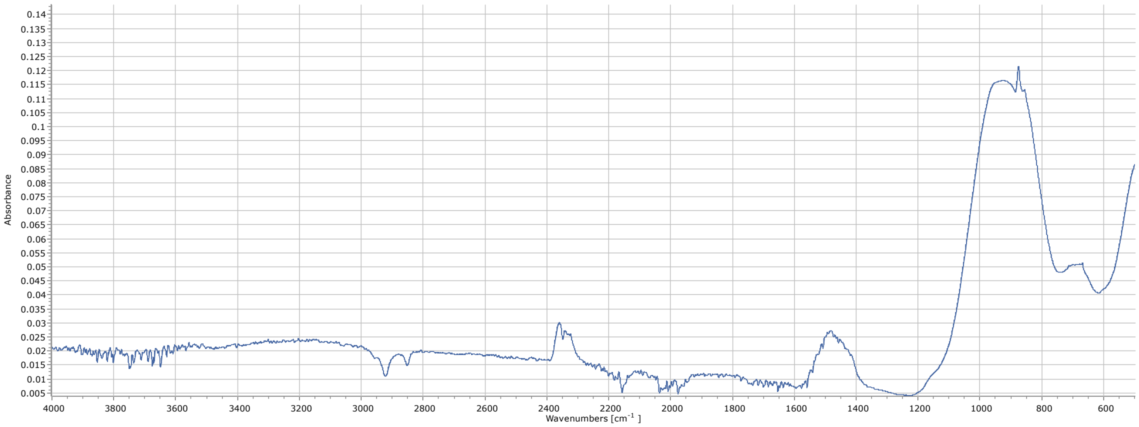

Fig. 1: FTIR spectrum of GBBS sample.

3.2. Changes in pH and temperature

The temperature remained constant before and after adsorption, wherein the initial temperature was around 23.2°C (room temperature) while the final temperature did not exceed 23.5°C for all tested solutions. On the other hand, as the dosage of ground granulated blast furnace slag (GGBS) increases, the pH of the solution rises due to the hydrolysis of metal oxides present in the slag when mixed with water. This process, as described by Shi et al. (2022) and Sahu et al. (2023), leads to the formation of hydroxide ions (OH-) and metal cations, increasing pH [10, 13]. The metal cations remain in the solution and only precipitate at specific pH values.

Table 1: pH values for different MP sizes and GGBS dosage

	Particle size (µm)		
GGBS dose (g/L)	**125-150**	**250-300**	**425-500**
2	9.62	9.4	9.35
4	9.6	9.15	9.31
6	9.68	9.4	9.29
8	9.68	9.18	9.37
10	9.6	9.29	9.45

3.3 Removal of MPs

At all doses, GGBS was effective in removing at least 72% of MPs. At the highest adsorbent dose (10 g/L), approximately 92% of MPs were removed from the solution. The physical adsorption characteristics of GGBS are determined by its porosity, specific area, and electrostatic attraction between the GGBS and the adsorbate [10]. As the pH of the solution was not adjusted before the experiments, the initial pH of the solution was basic due to the hydrolysis of the oxides in the GGBS. The hydroxyl groups in the solution (OH^-) can form adsorption sites on the surface of the GGBS particles, forming negatively charged surfaces, and attracting cationic adsorbates or adsorbates with positive surface charge [10]. At lower pH values, the hydrogen ions (H^+) present in the solution can protonate the surface charges of the GGBS, forming positively charged adsorption sites instead [10, 14]. The MPs used in this study were virgin pristine PE pellets, which are neutral and do not possess any acid-base properties (i.e., no surface charge and do not lose or gain electrons) when immersed in water. The electrostatic attraction, therefore, could not have played a role in the removal of MPs in this study, but can play a significant role in the removal of actual MPs in wastewater. Weathered MPs, particularly PE, possess negative surface charges in the environment as degradation impacts the chemical properties of the polymers and can create new functional groups, such as ketones, that increase the polarity of the compounds [15]. As MPs are suspended particles, other chemical adsorption reactions, such formation of surface coordination complexes, ion exchange, and chemical precipitation, could not have occurred. The GGBS used in this study was powdered, so the adsorbate had higher porosity and more active sites available for adsorption [16]. GGBS can also form calcium silicate hydrate (CSH gel) when in water, which can effectively remove colloidal particles. Fig. 2 shows the values of qe at different slag doses while Fig. 3 shows the final equilibrium concentration at different slag doses. Though the equilibrium concentration decreased with increasing slag doses, the amount of adsorbate removed per gram of adsorbent (adsorption efficiency) decreased with increasing slag doses. There were also no significant changes in the qe values for the three different MP sizes, even though the number of MPs in each solution (items/L) for each MP sample used were different due to the different sizes used.

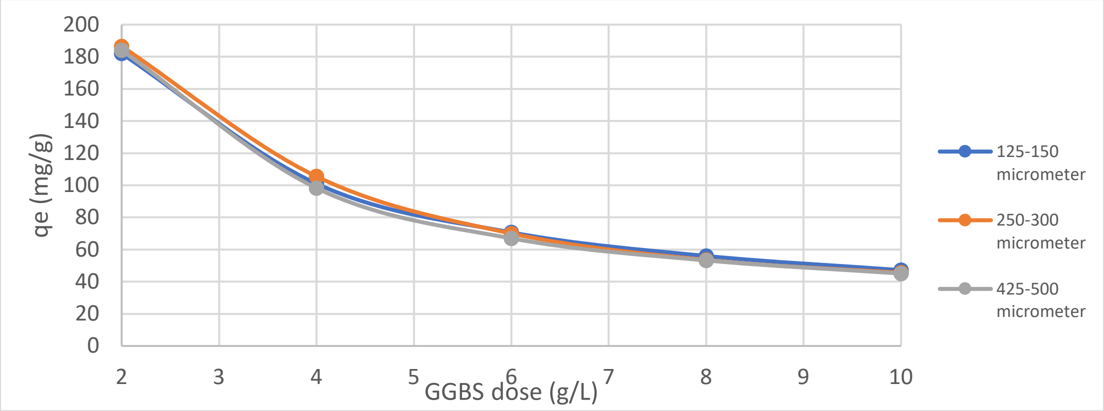

Fig. 2: Effect of GGBS on the adsorption density

3.3 Adsorption isotherms

Experimental results were applied to two commonly used isotherm models. The results show that the Freundlich isotherm is better suited for the adsorption of MPs onto GGBS, indicating that the adsorbent surface is non-homogenous and is multiphase [10]. The Langmuir isotherm displays poor R^2 values with negative qm and KL values, so the model can be dismissed. The dominant isotherm for adsorption on GGBS depends on the size of the adsorbent and the type of adsorbate. Generally, for GGBS particles smaller than 2 mm in size, the Langmuir isotherm provides a better-fitted model (Shi et al., 2022). In this study, however, the Freundlich isotherm was more suited. As there are no studies in the literature on the removal of MPs using GGBS, more studies are needed to determine the suitable isotherm.

Table 2 shows a summary of all the isotherm parameters from both the models. The n values for the Freundlich isotherm are less than 1 for MP sizes 250-300 micrometers ($1/n > 1$), indicating that adsorption is unfavorable for these MP sizes, despite the similar qe values reported for all three MP sizes [17]. More studies are needed to confirm these findings. If tested on weathered MPs instead of pristine samples, then the adsorption might be more favorable.

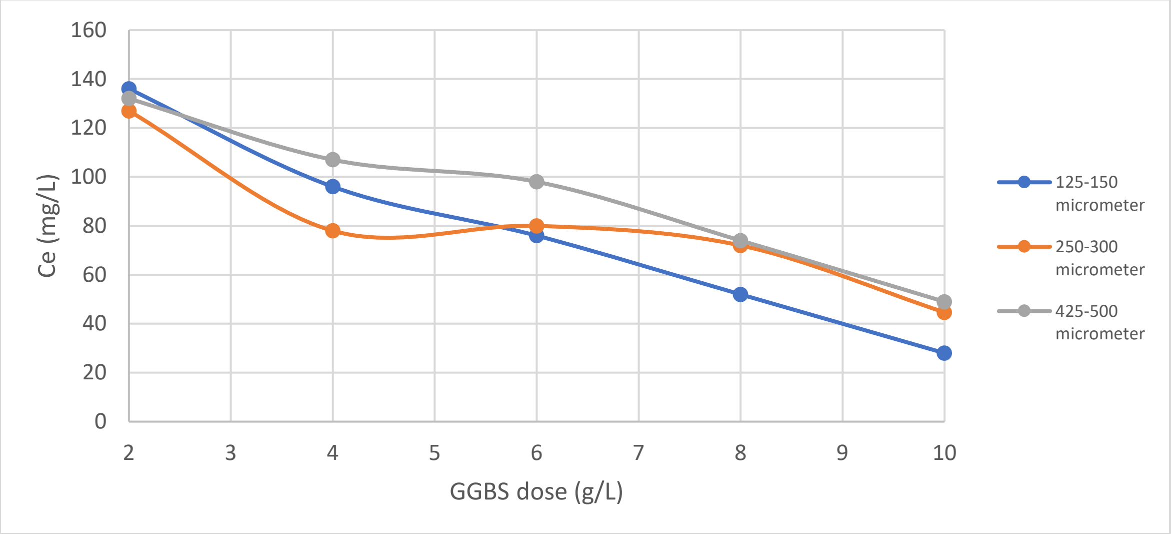

Fig. 3: Effect of GBBS on the equilibrium concentrations

Table 2: Isotherm parameters

Size (µm)	Langmiur			Freundlich		
	Q_{max} (mg/g)	K_L (L/mg)	R^2	K_f $(mg/L)^{-1/n}$	n	R^2
125-150	1111.11	1.14×10^{-3}	0.0393	2.5845	1.25	0.8753
250-300	-212.76	-3.47×10^{-3}	0.2520	0.2136	0.730	0.8907
425-500	-227.27	-2.82×10^{-3}	0.2275	0.2326	0.768	0.8127

3.4 Effectiveness of adsorption of MPs on GGBS

The results of this study show the potential of GGBS for the removal of MPs. The adsorption of MPs is not widely studied in the literature, despite the promising results and the viability of the treatment method. Wang et al. (2021) studied the efficiency of zing/magnesium magnetic (modified) biochar in the removal of MPs and reported suitable results, primarily due to the electrostatic attraction between the MPs and the modified biochar. The authors also reported that the metal oxide particles reinforced the adsorption of the MPs onto the adsorption sites available, enhancing the stability of the reaction [8]. For the removal of MPs, GGBS might not need to be modified as it already includes different metal oxides; instead, the batch adsorption tests can be carried out at acidic pH values (< 6) to ensure that the adsorption sites are protonated, which can attract the negatively charged weathered MPs.

Though the use of powdered GGBS provides greater surface area, which enhances the physical adsorption process—the main driver in this study—separating the powder from the solution was difficult, even after centrifuging, which affected the filtration process. During some experiments, the MPs immediately separated from the GGBS when pipetted, which could potentially indicate that the process is unstable, and that desorption is highly likely. Modifying the surface of the slag could potentially mitigate this problem through enhancing the electrostatic attraction between the GGBS and MPs. As MPs have a high affinity to metals, enhancing the GGBS surface with metals such as zinc could potentially improve the process further. Desorption experiments were not conducted in this study but should be take into account in future studies to measure the effectiveness of the process.

4. Conclusion

This study investigated the performance of batch adsorption tests to investigate the potential and kinetics of GGBS in adsorbing MPS. Three different average particle sizes, where each had a different particle, color were investigated: 125-150 micrometers (blue), 250-300 micrometers (red), and 425-500 micrometers (green). The results showed that at all doses, GGBS was effective in removing at least 72% of MPs. Indicating a potential for the use of slag to remove MPs from the environment. The ability GGBS to remove colloidal particles is attributed to the formation of CSH gel. This gel has adsorptive properties that enable the removal of colloidal particles from water. The formation of flocs due to the interaction between the CSH gel and colloidal particles contributes to the agglomeration and subsequent settling of the particles, leading to their removal from the water [18].

Future research could focus on further understanding the mechanisms underlying the adsorption of MPs by GGBS, as well as optimizing the conditions for maximizing its effectiveness in water treatment processes. Exploring the influence of factors like temperature, pH and plastic degradation on the adsorption capabilities of GGBS for microplastics is crucial. These factors can greatly impact the efficiency and cost effectiveness of removing microplastics. Another possibility would be considering using a larger size of GGBS for better separation of GGBS from the solution. However, a comprehensive study is required to assess any potential decrease in efficiency. Additionally, investigating the potential of GGBS in removing microplastics from different types of wastewaters, such as municipal, industrial, and agricultural wastewater could be valuable for practical applications.

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