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Microwave-Assisted Rapid Synthesis of C@Fe_3O_4 Composite for Removal of Microplastics from Drinking Water

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Abstract

Filtration is a basic requirement for the production of clean drinking water. However, filtering of large-scale drinking water is a time-consuming and costly process. Addressed herein is a new approach to the removal of microplastics, defined as dangerous organic pollutants, from water. As magnetic adsorbent, highly porous and well dispersed C@Fe_3O_4 composites were produced by a facile and rapid one-pot microwave synthesis method in minutes. The prepared C@Fe_3O_4 composites were used as an adsorbent in water contaminated with microplastics. The obtained results revealed that the microplastics adhered to the composite surface and were successfully removed from the water with an external magnet. In this point, this study provides a new approach to the rapid, effective, and low-cost removal of microplastic pollutants from drinking water.

Keywords: Filtration; Magnetic composites; Microwave-assisted synthesis; Micropollutant.

İçme Suyundan Mikroplastiklerin Uzaklaştırılması İçin C@Fe_3O_4 Kompozitinin Mikrodalga Destekli Hızlı Sentezi

Öz

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Filtrasyon, temiz içme suyu için temel bir gerekliliktir. Bununla beraber, büyük miktarda içme suyunun filtrelenmesi zaman alıcı ve maliyetli bir işlemdir. Tehlikeli bir organik kirletici olarak tanımlanan mikroplastiklerin sudan uzaklaştırılmasına yeni bir yaklaşım sunulmaktadır. Manyetik adsorban olarak, son derece gözenekli ve iyi dağılmış C@Fe_3O_4 kompoziti dakikalar içinde kolay ve hızlı mikrodalga destekli sentez yöntemiyle üretildi. Hazırlanan C@Fe_3O_4 kompoziti, mikroplastiklerle kirletilmiş suda bir adsorban olarak kullanıldı. Elde edilen sonuçlar, mikroplastiklerin kompozit yüzeye yapıştığını ve harici bir mıknatısla sudan başarıyla çıkarıldığını ortaya koydu. Bu noktada, bu çalışma mikroplastik kirleticilerin içme suyundan hızlı, etkili ve düşük maliyetli olarak uzaklaştırılmasına yeni bir yaklaşım sunmaktadır.

Anahtar Kelimeler: Filtrasyon; Manyetik kompozitler; Mikrodalga destekli sentez; Mikrokirletici.

1. Introduction

In recent years, magnetic materials have attracted a great attention because they can be produced effectively and efficiently from abundant raw materials [1, 2]. These materials can be used in different roles such as absorbents, catalysts, capacitors, energy storage systems, and sensors in various applications [3-9]. Especially, magnetic composites are used in the field of catalytic decomposition and removal of organic pollutants in environmental applications [10-12]. In addition, magnetic property significantly reduces recovery costs after the use of the material [10, 13]. This advantage allows the repeated use of these materials in many applications such as catalytic process and filtering systems [5, 14-16]. As a result of these advantages, magnetic materials will become more important in the future in terms of their potential applications. Therefore, the production of their composites in a cheap, fast, and an efficient way plays a vital role.

Microwave-assisted synthesis technology has been emerging as an alternative method for higher efficiency, selectivity, and large-scale production of nanoparticles compared to the conventional synthesis methods [17-23]. This technology is a fast synthesis technique that consumes relatively low energy [24, 25]. In particular, it provides efficient heating, which leads to a uniform size distribution of nanoparticles [23, 26, 27]. Thanks to controlled heating, it prevents the formation of many side reactions, and thus provides efficiency and repeatability in the production of nanoparticles [28-30]. Therefore, microwave technology is an indispensable part of green chemistry [19, 31].

The low-cost and fast production of magnetic composites have gained increasing interest as filtering materials to obtain clean drinking water [32, 33]. Carbon-based magnetic materials

are used to remove heavy metal ions, organic dyes, and microbial contaminants from water [34, 35]. In addition, microplastics are defined as organic pollutants that are becoming increasingly dangerous to human health [36-38]. New composites are being developed as an alternative to traditional filter materials such as natural minerals, zeolites, and porous carbon absorbents used to clean drinking water [39-41]. Due to its porous structure and high surface area, carbon-based magnetic composites can be an alternative to conventional filter materials to remove microplastic pollutants from drinking water [42, 43].

The present paper introduces a rapid synthetic strategy, which allows the production of highly porous and well dispersed C@Fe_3O_4 composites in minutes. Furthermore, the produced C@Fe_3O_4 composites were evaluated to remove microplastic pollutants from drinking water.

2. Experimental

2.1. Chemicals and materials

The reagent grade chemicals, Polystyrene beads (3μm), Glucose, $FeCl_3$, $FeCl_2$. $4H_2O$ and Ammonium hydroxide (30-32%) were purchased from Sigma-Aldrich. All reactions were carried out using deionized water (resistivity < 18 MΩ·cm).

2.2. Instrumentation

The TGA analysis was carried out with a HITACHI SII 7300 with a heating rate of 2 °C min^{-1} under an air atmosphere. The XRD (X-ray diffraction) patterns were performed on a Pan Analytical Empyrean instrument with Cu Kα radiation (λ = 1.54056 Å) from 3 to 70° (2θ) at a scanning rate of 2° min^{-1}. The micro-morphology and structure of sample were examined using Scanning electron microscopy, and Transmission electron microscopy (SEM, ZEISS Sigma 300; TEM, Hitachi HT 7700). The magnetic properties were performed on a VSM device (Quantum Designed Physical Property Measurement System) in the magnetic field range of ±20 kOe. The Brunauer−Emmett−Teller (BET) specific surface area of C@Fe_3O_4 was calculated by nitrogen sorption isotherms that were measured on a Micromeritics 3Flex instrument, to obtain the surface area. Microwave-irradiated reactions were conducted on a microwave reactor (Discover SP, CEM, Matthews, NC, USA).

2.3. Preparation of C@Fe_3O_4 composite

1.02 g of $FeCl_3$, 2.45 g of $FeCl_2$. $4H_2O$ was dissolved in a 80 mL of de-ionized water, and then 5 mL of 0.1M NH_4OH solution added over it dropwise. The solution was refluxed under microwave irradiation according to the desired program (200 W, 100 °C, hold for 10 min). 2 g of

glucose was dissolved in 5 mL of de-ionized water, then glucose solution added over as-prepared Fe_3O_4 suspension. The suspension was refluxed under microwave irradiation at 130 °C for 10 min. Finally, the C@Fe_3O_4 composite was filtered, washed with deionized water several times, and then dried at 70°C for 12 h.

2.4. Micropollutant removal batch studies

Drinking water samples were obtained from the local market in Turkey and used without further purification or any treatments in the micropollutant removal studies. In a sample batch study, an appropriate amount of polystyrene beads were dispersed in the drinking water for 30 min in an ultrasonic bath. Then, the resulting suspensions were treated with the produced C@Fe_3O_4 composites in order evaluate their micropollutant removal efficiencies from the drinking water samples.

3. Results and Discussion

Powder XRD was used to confirm the crystal structure of C@Fe_3O_4 composite. Fig. 1a shows the XRD diffraction pattern of C@Fe_3O_4.The X-ray diffraction pattern of the C@Fe_3O_4 exhibits a broad peak centered at 22.21°, which is attributed to amorphous carbon support. The peaks of Fe_3O_4 at 30.28°, 35.63°, 36.754°, 43.36°, 53.93°, 57.942°, and 63.58° are consistent with the (220), (311), (222), (400), (422), (511), and (440) of the standard card of Fe_3O_4 (JCPDS 65–3107) (Fig. 1a). XRD data confirms the presence of both magnetic Fe_3O_4 and amorphous carbon in the composite structure.

Separation of the produced composite from the suspended solution with an external magnet is an important feature to reduce recovery costs and saving time. Thus, synthesis of magnetite, which exhibit relatively high magnetization among other magnetic metal oxides, was chosen. The magnetic properties of C@Fe_3O_4 composite were investigated using VSM device (Quantum Designed Physical Property Measurement System). Fig. 1b display a representative magnetization curve of C@Fe_3O_4 measured at room temperature, which exhibit typical superparamagnetic behavior and the saturation magnetization of the C@Fe_3O_4 is 48.3 emu/g.

The solution containing Fe^{2+} and Fe^{3+} salts forms the highly soluble ferrihydrite ($Fe(OH)_3$) in basic medium [44]. It is then possible that $Fe(OH)_3$ can react with Fe^{2+} species and precipitate as magnetite (Fe_3O_4). The following reaction takes place (predictably) during microwave-assisted synthesis; $2Fe(OH)_3 + Fe^{2+} \rightarrow Fe_3O_4 + 2H_2O + 2H^+$.

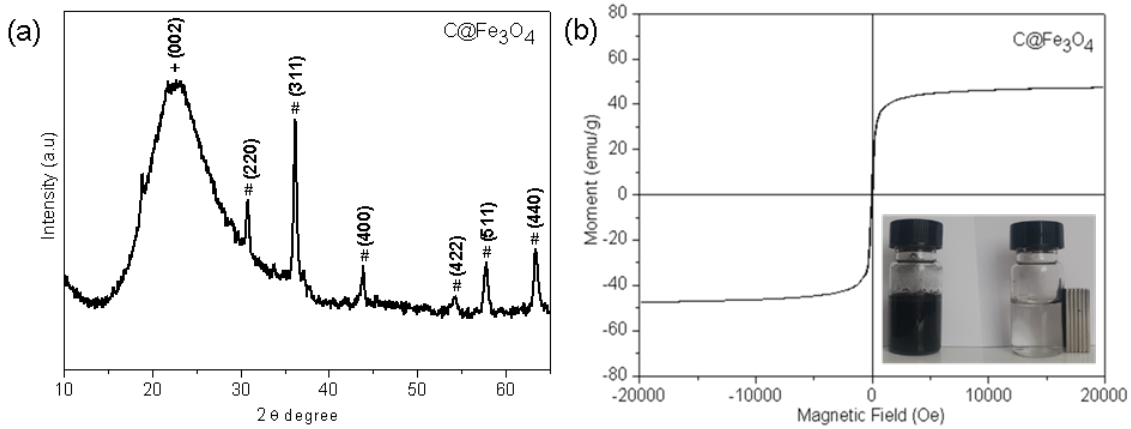
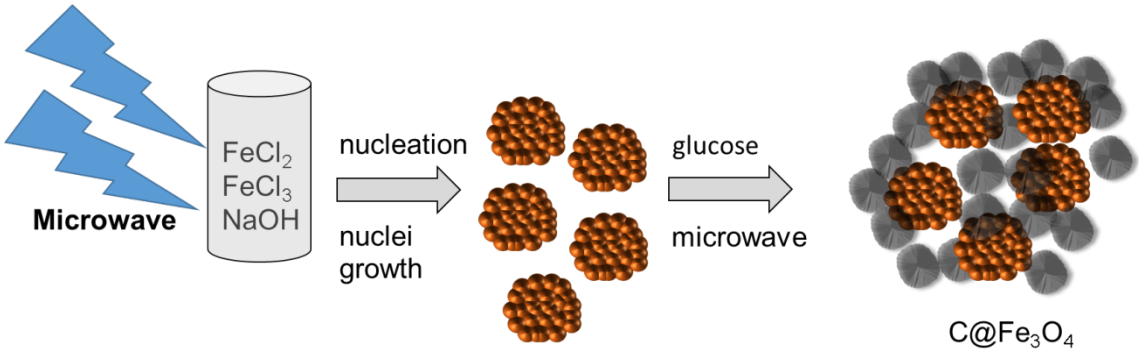


Figure 1: (a) XRD pattern of C@Fe_3O_4, (b) room temperature magnetization curve of C@Fe_3O_4

Thanks to the microwave-assisted controlled heating, this suspension was completely transformed into the nearly monodisperse Fe_3O_4 nanoparticles with a uniform size within a short period of 10 min (Scheme 1).



Scheme 1: Schematic of the synthesis procedure of C@Fe_3O_4

SEM and TEM analysis techniques were used to investigate the shape and size of the particles forming the components of the composite. SEM image of Fe_3O_4 particles in the composite revealed that the average diameter was ~90 nm, producing an interconnected porous network with carbon support seen in Fig. 2a. In addition, TEM image shows that carbon support has a spherical shape with an average size of ≈40 nm decorated with spherical Fe_3O_4 (Fig. 2b).

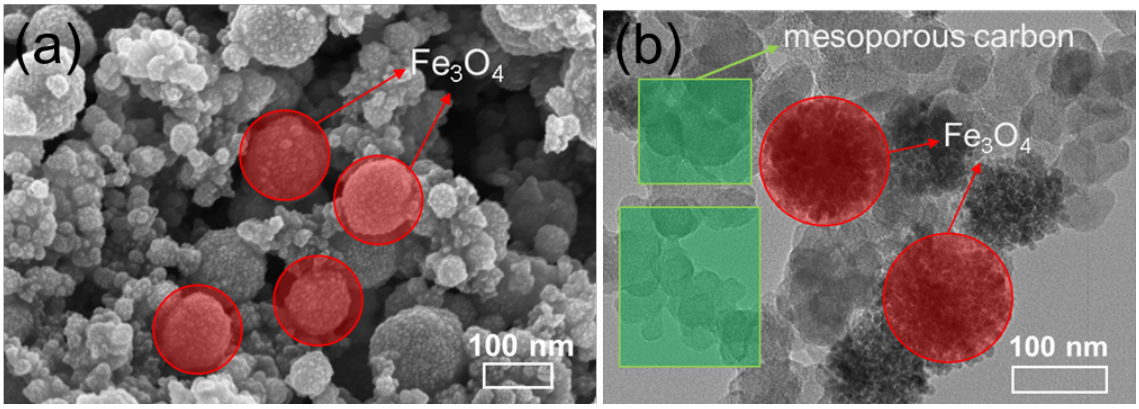


Figure 2: (a) SEM image of C@Fe_3O_4, (b)TEM image of C@Fe_3O_4

Thermal stability of the C@Fe_3O_4 was characterized by TGA (Fig. 3a). The first weight-loss in the temperature range of ca. 30−120 °C might result from the loss of adsorbed water. The second weight-loss broader range of ca. 250−450 °C was presumably due to the decomposition of the C@Fe_3O_4, in which the carbon support oxidized to CO_2. This is followed by a more drastic weight loss starting at ca. 450 °C, revealing a nearly complete decomposition of the Fe_3O_4 to Fe_2O_3.On the other hand, the porosity information on C@Fe_3O_4 composite was characterized by a N_2 adsorption-desorption study, and the BET surface area of C@Fe_3O_4 was measured to be 250.1 m^2/g.

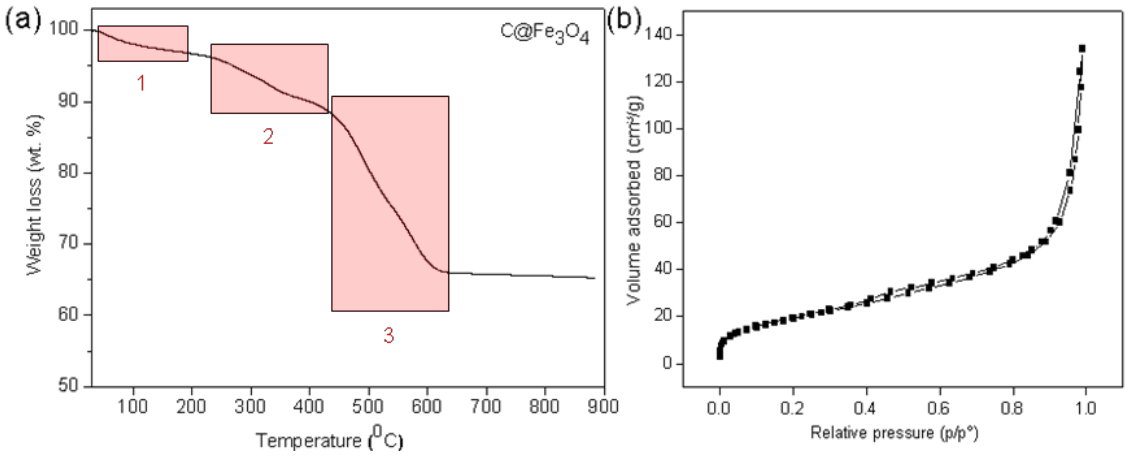


Figure 3: (a) TGA curve of C@Fe_3O_4, (b)Nitrogen adsorption-desorption isotherm of C@Fe_3O_4

Microplastics are defined among common aquatic pollutants with a size range of 0.1 µm - 5 mm [45, 46]. The main source of these pollutants is the result of mechanical and chemical decomposition of large quantities of plastics produced. Microplastics transported to the seas and lakes by wastewater threaten all aquatic life [47]. In addition, these pollutants are also a great threat to mammals through the food chain. Moreover, its toxicity to living organism in the

growing period is even higher [45-47]. Therefore, these organic pollutants should be removed from water sources with effective and low-cost methods. For this purpose, C@Fe_3O_4 composite was produced as a model sorbent by microwave-assisted synthesis method to remove microplastics from water. Polystyrene beads were used to represent microplastics as organic pollutants.

20 mg of polystyrene beads were dispersed in 20 ml of water for 30 minutes in the sonicator. Then 5 mg of composite was added to this suspension and the heterogeneous mixture was sonicated for 30 minutes. The suspension containing composite and polystyrene beads were separated by an external magnet from the drinking water in 2 minutes (Fig. 4b). Then, this structure was examined in detail with SEM and EDX measurements. SEM image shows that the C@Fe_3O_4 composite adheres to the surface of polystyrene beads (Fig. 4a). In this way, polymer beads suspended in water were easily separated with the help of an external magnet (Fig. 4b). Furthermore, the energy dispersive X-ray (EDX) elemental mapping clearly confirms that the C@Fe_3O_4 composite completely cover on the polystyrene beads (Fig. 4a).

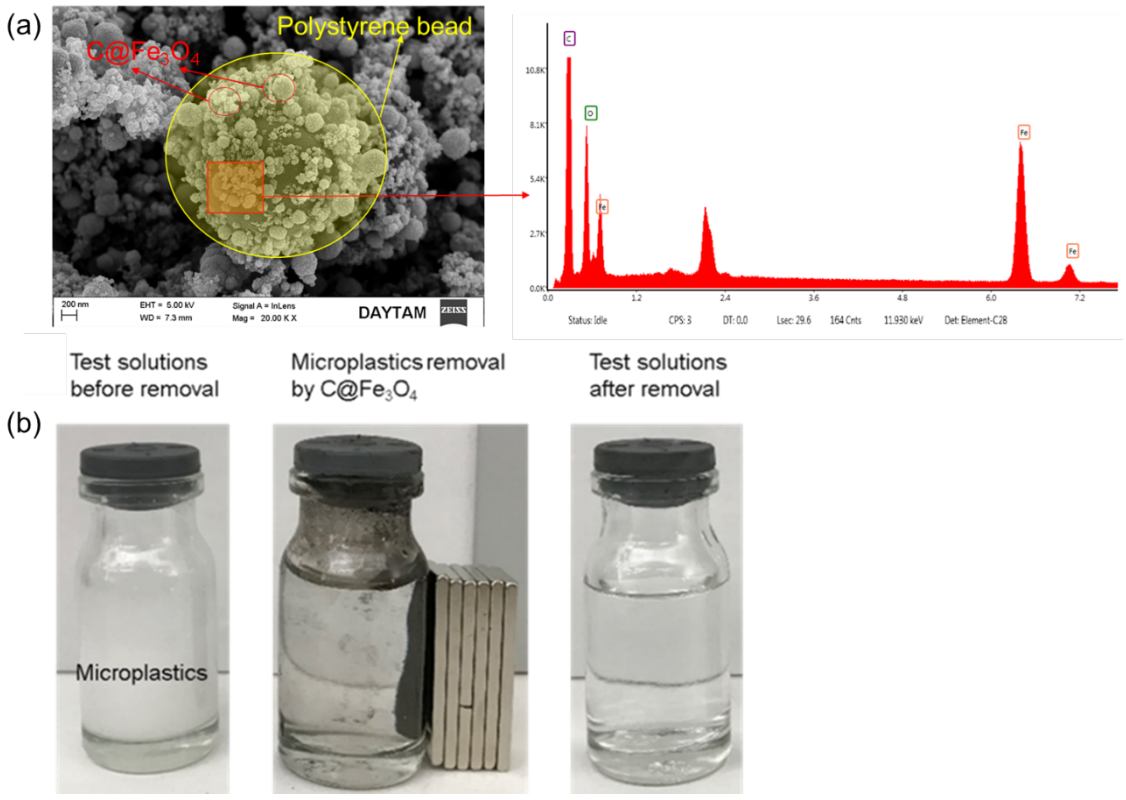


Figure 4: (a) SEM and EDX images of micro-sized polystyrene beads covered C@Fe_3O_4 composite, (b) Microplastics removal by magnetic C@Fe_3O_4. SEM image of micro-sized polystyrene beads covered C@Fe_3O_4 composite

4. Conclusions

In summary, we prepared highly porous and well dispersed C@Fe_3O_4 composite by a facile and rapid one-pot microwave synthesis method in minutes. The introduced microwave process led to the production of homogeneous particle size distribution Fe_3O_4 (~ 90nm) and high surface area carbon support (250.1 m^2/g), using low-cost starting materials. Furthermore, C@Fe_3O_4 composite was used as an adsorbent in water contaminated with microplastics. The microplastics adhered to the composite surface and were successfully removed from the water with an external magnet. Overall, this study provides a new approach to the rapid, effective, and low-cost removal of microplastic pollutants from drinking water samples.

Acknowledgement

The author declares no conflict of interests.

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