



## Gel-emulsion templated polymeric monoliths for efficient removal of particulate matters



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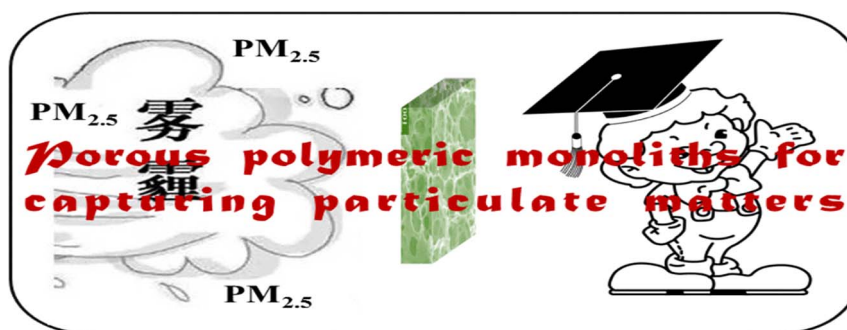
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### HIGHLIGHTS

- Acidified cotton is used as an efficient stabilizer of W/O gel-emulsions.
- Porous polymeric monoliths are produced from the templates.
- The densities of the monoliths could be as low as 0.06 g/cm<sup>3</sup>.
- Particulate matters can be efficiently removed by using the monoliths.

### GRAPHICAL ABSTRACT



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### ABSTRACT

In this work, a series of porous polymeric monoliths with densities as low as  $\sim 0.060$  g/cm<sup>3</sup> were prepared by employing some newly created gel-emulsions as templates, of which acidified cotton was utilized for the first time as an efficient stabilizer. The monoliths as obtained could be purified by simple washing and dried at ambient conditions. Moreover, they showed strong affinity to particulate matters (PMs). Adsorption tests revealed that the saturated adsorption capacity of one of the monoliths (M8) to PM<sub>2.5</sub> could reach  $\sim 520$  mg/g and the removal efficiency exceeds 93% at ambient conditions, a prominent result considering the collective advantages of the monolith. Importantly, the monoliths after adsorption can be recovered and re-used via simple washing and drying. More importantly, the stabilizer as created is so efficient that 0.1% (w/v, cotton to the volume of the continuous phase) is enough to stabilize the gel emulsions. Considering the excellence in performance, low cost in preparation, and convenience in regeneration, we believe that the monoliths are strong candidates for real-life applications.

### 1. Introduction

Particulate matter (PM) pollution has recently become one of the most serious environmental issues that impose huge threat to public health [1,2]. PM is a complex mixture of extremely small particles and

liquid droplets. It can be categorized into PM<sub>2.5</sub> and PM<sub>10</sub>, which refer to particle sizes below 2.5 and 10  $\mu$ m, respectively [3]. What is more, PM<sub>2.5</sub> is particularly harmful since it can penetrate human bronchi and lungs owing to its small size. Hence, long-term exposure to PM pollution can lead to heart disease, strokes, and lung disease, including

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cancer [4–7], while short-term exposure to high concentration of PM can trigger asthma attacks [8]. Due to its severe impact on human health, it is necessary and urgent to deploy effective protection for the public from present hazy days [9]. Therefore, development of efficient technology for removal of PM is strongly needed.

Principally, there are four strategies for removing the pollutant from air: the first is chemical reduction [10], the second chemical catalytic oxidation [11], the third physical filtration [12–16], and the last physical adsorption [17–19]. Although the methods have made some progress, they are still far from satisfied due to reasons such as low efficiency, high cost, and complicated operation, etc. Compared with chemical approaches, physical methods inherently possess several advantages, such as: 1) easier to be implemented, 2) less energy consumption, and 3) generally reusable.

The key point for physical methods as mentioned is the development of high performance adsorbents. To date, widely studied physical adsorbents mainly include ceramics [20], polyvinyl chloride/polyurethane fibers [21], sandwich structured polyamide-6/polyacrylonitrile nanonets/bead-on-string composite membranes [22], polytetrafluoro-ethylene nanoparticles [23], etc. These materials have been getting more and more attention over the last few years owing to their superior filtration and adsorption efficiencies if compared with conventional air cleaning methods. Moreover, they offer high-temperature filtration capability, corrosion resistance, and durability. However, these materials still have their own limitations, such as difficult in regeneration, complicated in preparation, expensive in cost, and in particular, requiring large space in using.

In a previous report [24], we demonstrated that porous polymeric monoliths such as polystyrene-divinyl benzene (PS-DVB) showed much stronger affinity to volatile organic compounds (VOCs) than most of the adsorbents reported in literatures [25]. Partial of the reasons for the high efficiency is their porous internal structures. Considering the key point for an ideal PM adsorbent is its optimized internal structure that is relatively large specific surface and interconnected networked channels [26], the porous polymeric monoliths as afore mentioned are expected to be good adsorbents for removing PM from air. Moreover, the porosity may also endow the materials superior adsorption/desorption kinetics, which is also crucial for practical uses [27–30].

Practically, the porous materials are generally prepared by using colloids or other dispersion systems as templates. In a typical preparation, a stable biphasic system is generated first, and then the continuous phase is polymerized. The colloidal entities serve to create porosity in the final polymeric monoliths. Among the templates, however, gel emulsions are more unique as their internal structures and properties can be easily adjusted by varying their compositions of continuous phase and content of dispersed phase [31], realizing fine control of their structures from highly interconnected to isolated networked cells.

As well known, the volume fraction of the dispersed phase of a conventional gel-emulsion must be greater than 74%, which is a critical value in geometry for a container to be fully filled by non-deformed balls [32]. Moreover, to be gel-emulsions, a third component must be introduced to stabilize the dispersion via minimizing the interface energy of the system. This component is named as the stabilizer of gel-emulsions [33].

A few years ago, our group reported for the first time to use low-molecular mass gelators (LMMGs) as efficient stabilizers [34–36]. Different from conventional ones, these gel-emulsions could be generated with the volume fraction of the dispersed phase much lower than 74%, greatly expanded the scope of volume fraction of dispersed phase, laying foundation for enhancing their template applications [37]. However, the LMMGs have to be designed and synthesized, which are generally tedious and complicated, and significantly elevate the cost of the materials. To our lucky, we found for the first time that the suspension from concentrated sulfuric acid treated cotton could be used as an efficient gelator of the mixture of styrene (St), divinyl-benzene

(DVB) and water, a typical water in oil (W/O) gel-emulsions. The gel-emulsions as obtained were further used as templates for preparing low-density porous polymeric monoliths and even aerogels in this case. This paper reports the details.

## 2. Experimental section

### 2.1. Materials

Cotton was purchased from Tansoole and used without further purification. DC1700 Air Quality Monitor is a product of USA (Patent: 8009290). Winner 311XP spray laser granulometer is a product of Jinan Weina Particle Instruments CO., Ltd. Differential pressure gauge (SNDWAY, SW512) was purchased from Guangdong SNDWAY Instruments CO., Ltd. Sulfuric acid (98%) was purchased from Sinopharm Chemical Reagent CO., Ltd. Styrene (St) and divinyl-benzene (DVB) are of chemical pure, and products of TCI. They were purified by passing through a basic alumina column before use to remove the pre-added inhibitor. 2,2'-Azobis(isobutyronitrile) (AIBN, 97%) was purchased from Sigma-Aldrich and used directly without further purification. Hydrophobic SiO<sub>2</sub> and *n*-octyltriethoxy-silane (*n*-OTES) were purchased from Aladdin. All solvents used in the studies were purified in the way as described in solvents handbooks. Water used throughout was doubly distilled. Other reagents, except those specially indicated, were used as received.

### 2.2. Preparation of gel-emulsion

A certain amount of cotton (25 mg) were firstly dissolved in 2.5 mL concentrated sulfuric acid (98%). The transparent solution as generated was used as a new type of stabilizer. In a typical preparation, the oil or the continuous phase was prepared in a test tube, which is composed of AIBN, St, DVB, *n*-OTES, SiO<sub>2</sub> and a certain amount of afore mentioned stabilizer. Then, a suitable amount of water was added to the oil phase. The mixture as obtained was intensely stirred or vigorously shaken for a while. In this way, a gel emulsion can be generated. The formation of a gel-emulsion was confirmed by inverting the test tube to observe if the mixture inside could still flow or not. The compositions of the gel-emulsions are listed in Table 1.

### 2.3. Characterization of the Gel-emulsion

#### 2.3.1. Fluorescence microscopy observation

Confocal laser scanning fluorescence microscopic image of the gel emulsion was taken on a TCS SP5 laser scanning confocal microscopy spectrometer. The probe used in the measurement is a known derivative of perylene bisimide, of which the structure is shown in Scheme S1. The excitation and emission wavelengths employed are 365 nm and 550 nm, respectively.

#### 2.3.2. Rheological measurements

Rheological measurements were carried out with a stress-controlled rheometer (TA Instruments AR-G2) equipped with a steel-coated parallel-plate geometry (20 mm diameter). The gap distance was fixed at 1000 μm. A solvent-trapping device was placed above the plate to avoid evaporation. All measurements were conducted at room temperature (20 °C). A stress sweep measurement at a fixed frequency (1.0 Hz or 6.28 rad/s) was conducted to explore storage modulus (*G'*) and dynamic yield values of gel emulsion, which provides information about the mechanical strength of the gel sample.

### 2.4. Preparation of porous polymeric monoliths

Gel-emulsions as obtained were thoroughly degassed, then heated to 50 °C, and then maintained at the temperature for 4 h to start pre-polymerization. The temperature of the system was raised to 75 °C and

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