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Ultra-rapid assembly of graphene oxide macroscopic materials enabled by low-surface-tension solvent



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ABSTRACT

Two-dimensional (2D) nanomaterials assembled macroscopic materials via bottom-up approach are extensively utilized across various domains, attributable to their capacity to retain the desirable properties of individual nanosheets. However, the dense structure formed during assembly restricts solvent molecule passage, leading to prolonged preparation times. Traditional methods struggle to create larger and more solvent channels. Herein, we propose a strategy to achieve ultra-fast assembly of graphene oxide (GO) nanosheets by manipulating the solvent's surface tension to form a bouffant structure. This approach enables GO membrane assembly in only 10 s, which is 24,660 times faster than that of conventional water-based methods (over 2.85 days). Our analysis of solvents with varying surface tensions demonstrates that low surface tension, rather than solvent's relative molecular weight, significantly impacts the assembly rate. This rapid assembly process enables GO membranes to attain thicknesses of several centimeters, thereby overcoming the limitations of traditional bottom-up techniques. The resulting GO macro-materials exhibit considerable potential for diverse applications, from thin membranes for waste management to thick blocks for electromagnetic interference shielding and mechanical sensors. This work offers valuable insights into the rapid assembly of macroscopic structures from GO and potentially other 2D lamellar materials.

1. Introduction

The assembly of promising two-dimensional (2D) nanomaterials into three-dimensional (3D) macroscopic structures with customized architectures or novel functionalities holds practical importance [1]. Graphene oxide (GO) macroscopic assemblies including membranes, demonstrate promising applications in seawater desalination [2], organic solvent nanofiltration [3], sensors [4], energy storage [5], etc. In practical applications, GO macroscopic assembly membranes must be fabricated efficiently and with scalable thicknesses to meet diverse operational requirements [6]. However, the narrow nanochannels in GO membranes with tightly stacked structure, significantly limit solvent permeance [7–9], resulting in prolonged assembly times, which may extend up to approximately one week [10]. Furthermore, according to the Hagen-Poiseuille equation [11,12], membrane permeance is proportional to the square of the effective pore radius and inversely proportional to the thickness. Therefore, in the filtration-based macro-assembly of GO membranes, it is challenging to simultaneously achieve both a rapid macro-assembly rate and substantial membrane thickness.

Current techniques, such as bubble-facilitated assembly, have been employed to promote the assembly of GO membranes [13], but inevitably introduce complexity into the experimental procedures. Increasing the interlayer spacing of GO membranes is a widely adopted strategy to enhance permeance, shorten assembly time and increase membrane thickness [14]. For instance, Liu et al. reported that TiO₂ intercalation

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promoted an increase in interlayer spacing and solvent permeance within GO membranes [15]. Additionally, controlling the lateral size of GO nanosheets and engineering the pores can shorten the transport pathways of solvent molecules and expedite membrane preparation processes [16]. Porous GO membranes prepared with large interlayer spacing are expected to exhibit reduced assembly times and enhanced solvent permeance [17–19]. Nonetheless, the expansion of interlayer spacing has a limited effect, and the membrane-forming rate exhibits an exponential decline with increasing GO membrane thickness.

In recent years, researches have demonstrated that the interaction between GO nanosheets and solvent molecules is critical in facilitating solvent transport within membranes and influencing the assembly rate of GO membranes [13,20]. Zhao et al. proposed a strategy to rapidly assemble GO nanosheets into highly ordered membranes at the incompatible hydrogel/oil interface induced by shear force [21]. By incorporating a super-hydrophilic interlaminar material within the GO interlayer, our team achieved the ultrafast assembly of GO membranes in just 18 s [22]. However, these strategies still necessitate sophisticated material synthesis processes, which impede their further development. Consequently, developing a straightforward and rapid assembly strategy for macroscopic GO membranes remains a significant challenge.

In this work, we propose an efficient approach to create larger and more solvent channels by forming a bouffant architecture through a lowsurface-tension solvent, facilitating ultra-rapid macroscopic assembly of GO membranes in only 10 s, which is 24,660 times faster than that of traditional methods employing water as the solvent (over 2.85 days). Benefiting from this rapid assembly rate, the thickness of the GO membrane can be precisely controlled and adjusted from the nanometer scale to the centimeter scale, effectively addressing the trade-off between assembly rate and thickness scalability. The thickness-adjustable GO membrane exhibits significant potential in applications ranging from organic solvent nanofiltration with thin membranes to electromagnetic interference shielding and pressure sensors with thick blocks. We assert that this work offers valuable insights into the ultrafast assembly of macroscopic architectures based on GO and potentially other 2D lamellar materials.

2. Experimental

2.1. Preparation of GO@solvent membranes

Graphene oxide (GO) purchased from Wuxi Chengyi Education Technology Co., Ltd., was added into 25 mL various solvents at a concentration of 1.5 mg/mL, including water, methanol, ethanol, propanol, isopropanol, butanol, N-methyl-2-pyrrolidone (NMP), N,Ndimethylformamide (DMF), acetic acid, ethyl acetate. The mixtures were stirred evenly in a homogenization mixer for 20 min and subsequently vacuum-filtered through PTFE filter paper (pore diameter = $0.45 \,\mu\text{m}$, diameter $\Phi = 4 \,\text{cm}$). GO membranes were maintained in a wet state after vacuum filtration to evaluate solvent permeation and dye rejection.

2.2. Preparation of GO@methanol materials with different thicknesses

By adding 0.15, 1.5, 1.2, and 20 mg/mL of GO to 10, 25, 150, and 150 mL of methanol solvent, respectively, followed by dispersion for 10, 20, 120, and 120 min, and subsequent vacuum filtration using PTFE filter paper (pore size = 0.45μ m, diameter $\Phi = 4$ cm). Macroscopic graphene oxide materials with varying thicknesses of 250 nm, 64 μ m, 5 mm, and 1.3 cm can be prepared.

2.3. Measurements of solvent permeation and dye rejection rate

A vacuum filtration apparatus was employed to quantify solvent permeation across the fabricated GO membranes under controlled pressures ranging from 0 to 1 bar. The contact area between GO@methanol membrane and the feed solution (solvent or dye solution) is 12.57 cm² (equivalent to a circular area with a diameter of 4 cm). The permeance is calculated using the formula $J = V/(A \times t \times P)$, where J, V, A, t, and P denotes the solvent's permeance (L m⁻² h⁻¹ bar⁻¹), the volume of permeance (L), the effective area (m²), the permeance time (h), and the applied pressure (bar), respectively. UV/Vis absorption spectra was employed to evaluate dye retention rates and quantify the concentrations of six dyes, including methylene blue (MB), methyl orange (MO), orange II (OG), Sudan I (Su I), rhodamine-6G (RG), butyl rhodamine (BRh) in the filtrate. Specifically, the dye retention rate was assessed using the formula $\eta = (1-C_1/C_0) \times 100\%$, where η , C_1 , and C_0 represents the dye retention rate (%), the dye concentration in the filtrate (µmol/L), respectively.

2.4. Preparation of RGO@methanol pressure sensors

GO was incorporated into 150 mL of methanol solvent at a concentration of 12 mg/mL. The mixture was stirred evenly in a homogenization mixer, followed by vacuum filtration to produce GO@methanol hydrogels. The resulting GO@methanol hydrogel structure was subjected to solvent exchange with water in a glass container, then freezedrying conducted in a lyophilizer. RGO@methanol aerogels were synthesized using a chemical reduction method and immersed in a solution blend (V_{47% hydriodic acid/Vethanol} = 1/3) for 12 h at room temperature. The RGO@methanol aerogels were then thoroughly washed with ethanol several times and dried overnight at 50 °C in an oven. Finally, an RGO@methanol pressure sensor was assembled by sandwiching a small section of RGO@methanol aerogel between two copper strips with a flag-shaped structure.

2.5. Sensitivity and stability evaluation of RGO@methanol pressure sensor

The RGO@methanol pressure sensor was positioned with attached copper tape within the fixture of universal testing machine (Instron 5882). Polyethylene terephthalate (PET) films were utilized to separate the contact between metal fixture in the universal testing machine and RGO@methanol pressure sensor to eliminate the impact of conductive fixture on the collected electrical signals. The fixture moved at a speed of 0.5 mm/min, and two computers were employed for pressure and electrical signal acquisition, respectively. Sensor sensitivity (*S*) is calculated using $S=\Delta_R/(R_0\times P)$, where *S* represents sensitivity of RGO@methanol pressure sensor (kPa⁻¹), Δ_R denotes resistance change (Ω), R_0 indicates initial resistance (Ω), and *P* signifies pressure (kPa). Sensor stability was assessed through cyclic pressure testing.

2.6. Electromagnetic shielding testing method

The electromagnetic interference shielding performance of RGO@methanol aerogel was assessed in a rectangular waveguide, utilizing a vector network analyzer (VNA, Keysight N5225A) within the X-band frequency range (8.2 - 12.4 GHz). Due to electromagnetic wave losses introduced by the circuits connected to the instrument, prior calibration and compensation are necessary to minimize the associated system errors. The RGO@methanol aerogel should be cut into rectangular pieces, slightly larger than the rectangular apertures in the waveguide chamber (10.2 mm \times 22.9 mm). During installation of the aerogel into the waveguide chamber, particular attention must be given to prevent any leakage at the edges, and the sample clamp should be firmly secured with screws.

3. Results and discussion

3.1. Ultrafast macroscopic assembly of GO membrane in low-surfacetension solvents

The bottom-up approach is a fundamental strategy for achieving the macroscopic assembly of two-dimensional (2D) nanomaterials, especially graphene oxide (GO) [23,24]. Among these methods for bottom-up assembly, vacuum filtration is extensively employed in the fabrication of GO membranes, enabling precise control over macroscopic dimensions and microstructural characteristics via the adjustment of process parameters [25]. GO can be assembled into macroscopic structures through hydrogen bonding, π - π stacking interactions, and electrostatic forces, among others [26]. The process involves the uniform dispersion of GO nanosheets in a solvent, followed by filtration under negative pressure to form a GO@solvent membrane (Fig. S1). We characterized the C/O ratio of graphene oxide powder by XPS to be 2.69 (Fig. S2). The vacuum filtration process comprises two stages: (i) the deposition of GO onto the filter paper surface under the vacuum-induced driving force, and (ii) solvent permeation through the deposited GO membrane [27,28]. During the solvent permeation stage, the solvent passes through the upper surface of the GO membrane, penetrates into its interlayer, and subsequently exits through the lower surface. However, the assembly time for GO membranes is typically prolonged, presenting a significant challenge to scaling up production. For instance, the preparation time for GO@water membranes with a thickness of 13 µm typically spans 2.85 days, as demonstrated in our practical benchmark experiments. During the assembly process, the capillary force (F_c) overcomes electrostatic repulsion (Fr), leading to the dense packing of GO flakes in water and the formation of narrow solvent channels (Fig. 1a) [29]. By regulating solvent polarity, solvents with lower

surface tension, such as methanol, induce the formation of a bouffant structure ($F_c < F_r$), thereby increasing and widening solvent channels. The hydrogen bonding strength of water (4.9 kcal mol⁻¹) [30], exceeds that of methanol (3.9 kcal mol⁻¹) [31]. The relatively weak hydrogen bonding ability of methanol reduces the existing hydrogen bonds between the GO layers, consequently decreasing the interlayer cross-linking density and fostering the formation of bouffant structures. This dynamic equilibrium facilitates the penetration of solvent molecules into the interlayer channels. thereby promoting the rapid assembly of GO macroscopic membranes (Fig. 1b). As illustrated in Fig. 1c and Video S1, the macroscopic assembly of the GO@methanol membrane occurs within 10 \pm 2 s, which is 24,660 times faster than the assembly of the GO@water membrane (2.85 \pm 0.08 days). The strength of the interaction between GO and the solvent directly influences the morphological characteristics of the membrane: a strong interaction leads to flat surface structures, whereas a weak interaction favors the formation of wrinkled structures. When the GO-solvent intermolecular interactions are weakened and become less than the threshold of the GO intramolecular interactions, thereby inducing the GO nanosheet to transform from flat extended state to wrinkled state [32].

Solvents with higher surface tension (e.g., water) generate stronger capillary forces, which promote the formation of a densely packed structure and a relatively smooth surface (Fig. $1d_1$ and $1e_1$) [33]. In contrast, solvents with lower surface tension (e.g., methanol) generate weaker capillary forces, which promote the formation of a bouffant structure and a relatively rough surface (Fig. $1d_2$ and $1e_2$) [34]. Scanning electron microscope (SEM) surface images (Fig. S3) also corroborates this significant difference, which are consistent with the structural features observed in super-depth three-dimensional microscope images shown in Fig. S4. The pore diameter distribution of the GO@methanol and GO@water membranes was analyzed via mercury intrusion



Fig. 1. Schematic of the fabrication procedure comparison between (a) GO@water membrane and (b) GO@methanol membrane (F_r = electrostatic repulsion, F_c = capillary force). (c) Membrane assembly time for GO@water membrane and GO@methanol membrane. Scanning electron microscopy (SEM) cross-sectional morphology image of (d₁) GO@water membrane and (d₂) GO@methanol membrane. White light interference images of (e₁) GO@water membrane and (e₂) GO@methanol membrane and GO@water membrane. (f) Pore diameter distribution of GO@methanol membrane and GO@water membrane.

porosimetry, as shown in Fig. 1f and Fig. S5, confirming the larger pore size of the GO@methanol membrane. Therefore, compared to GO@water membranes, GO@methanol membranes exhibit wider and more solvent channels, thereby providing additional spaces and pathways while reducing the number of tortuous routes for solvent molecules, and enhancing the assembly efficiency. Fig. S6 illustrates the measurement of the contact angle between water and methanol in the GO@water membrane, demonstrating that solvents with low surface tension, such as methanol, exhibit smaller contact angles, facilitating easier penetration. As shown in Fig. S7, the densities of the GO@water membrane and the GO@methanol membrane were determined to be 0.186 g/cm³ and 0.005 g/cm³, respectively.

3.2. The influence of GO membrane formation rate

Given that the assembly time of the GO@methanol membrane is

merely 10 s, while the settling time spans up to 3 min (Fig. S8, S9, and Table S1), it can be inferred that the aggregation of GO is not the primary factor driving the rapid assembly. Fig. S10 illustrates the Fourier transform infrared spectroscopy (FTIR) analysis of both the GO@methanol and GO@water membranes. The spectral feature observed at 1070 cm⁻¹ corresponds to the symmetric vibrational modes of the -CH₃ group, indicating specific resonance patterns associated with methanol. C=C stretching vibrations at 1630 cm⁻¹, C=O stretching vibrations at $1750 \sim 1720 \text{ cm}^{-1}$, C-O stretching vibrations at $1110 \sim 1040 \text{ cm}^{-1}$, and O-H stretching vibrations at $3340 \sim 3320 \text{ cm}^{-1}$ are observed [35], and these are attributed to the hydroxyl and carboxyl functional groups of GO. These findings suggest negligible differences between the prepared GO@methanol membrane and the conventional GO@water membrane. To further examine the primary factors influencing the assembly rate of GO membranes, a variety of solvents were investigated, including water, N-methyl-2-pyrrolidone (NMP), N,N-dimethylformamide (DMF).



Fig. 2. (a) The SEM of surface structure of GO@solvent (ethanol, propanol, DMF, and NMP) membranes. (b) Membrane formation rate and (c) permeance of GO membranes within different solvents (water, NMP, DMF, butanol, propanol, isopropanol, ethanol, acetic acid, ethyl acetate, and methanol). The influence of (d) surface tension and (e) relative molecular weight of solvent on membrane forming rate. The insets of (d) is self-supporting GO membrane in bending state.

butanol, propanol, isopropanol, ethanol, acetic acid, ethyl acetate, and methanol.

Among these, the surface (Fig. 2a) of GO membranes formed in ethanol, propanol, DMF, and NMP were primarily analyzed, demonstrating that the relationship between structural characteristics and assembly time corresponds to the patterns depicted in Fig. 1d, 1e and S2. Detailed comparisons of the assembly rates of GO dispersed in various solvents, along with their respective solvent permeance, are shown in Fig. 2b and Fig. 2c. The results indicate that the assembly rate of GO in different solvents follows a trend similar to the permeance of the GO membrane to the corresponding solvent post-assembly. Specifically, a higher assembly rate of GO in a given solvent is correlated with a higher permeance of the GO filter membrane in that solvent. Previous studies suggest that solvent permeance through the membrane depends on molecular mass (size) and pore structure of filter membrane [36,37]. In particular, solvents with smaller molecular masses (sizes), such as methanol, as well as more and larger pore structures of filter membrane, exhibit higher solvent permeance.

Fig. 2d examines the primary influence of solvent surface tension on assembly rates, showing that solvents with lower surface tension typically result in accelerated assembly rates, supported by a fitting curve with a goodness-of-fit of 92.55%. Some outliers deviating from the fitted curve suggest the involvement of additional factors influencing the membrane formation rate. For example, the surface tension of acetic acid exceeds that of ethyl acetate. Based on the trend predicted by the fitting formula, the assembly rate of GO@acetic acid is expected to be slower than that of GO@ethyl acetate. However, this prediction is inconsistent with the actual experimental results. As shown in Fig. 2e, this anomalous phenomenon is linked to the relative molecular weight of the solvent molecules. Smaller relative molecular weights of solvent improve solvent permeance and accelerate the assembly rates of GO membranes [38]. As depicted in **Fig. S11**, the methanol solvent permeance of the GO@methanol membrane attains a peak value of 14,905 \pm 194 L m⁻² bar⁻¹ h⁻¹. Moreover, **Fig. S12** illustrates a notable decline in methanol permeance as membrane thickness increases, which may be attributed to the extended methanol path length in thicker membranes.

3.3. Organic solvent nanofiltration performance of GO@methanol membrane

The GO@methanol membrane is suitable for dye treatment in solvent systems. Dyes, with an estimated annual production of approximately 700,000 tons, are pervasive in daily life, encompassing >100,000 commercial variants [39]. Specifically, organic dyes, including methylene blue (MB) and methyl orange (MO), are widely utilized across numerous industries, such as textiles, packaging, pharmaceuticals, and cosmetics [40]. Industrial wastewater containing organic dyes necessitates rigorous treatment before being discharged into natural water bodies, owing to the high toxicity associated with their complex aromatic ring structures and xenobiotic properties [41]. Six organic dves (Fig. S13), namely MB, MO, Sudan I (Su I), rhodamine 6G (RG), butyl rhodamine (BRh), and orange II (OG), were selected as representative object to investigate the interception properties of the GO@methanol membrane. Among them, MB, Su I, and RG are positively charged, while BRh is neutrally charged, and both MO and OG are negatively charged (Table S2).

The negatively charged GO lamella in GO@methanol membrane selectively intercepts positively charged dye molecules (e.g., MB, Su I, and RG) due to the predominant electrostatic attraction [42]. In contrast, electrostatic repulsion leads to low rejection rates for



Fig. 3. (a) Schematic diagram of GO@methanol intercepting dye molecules. (b) Separation performances of the GO@methanol membrane as demonstrated by various methanol solutions containing different organic dyes. (c) Comparison of methylene blue dye separation performance and methanol permeance of GO@methanol membrane and the reported membranes. (d) Methylene blue rejection and permeance of GO@methanol membrane under a compressive stress of 0.85 bar for up to 2 h.

negatively charged dyes (e.g., MO and OG), thereby facilitating their passage through the pores of the GO@methanol membrane with minimal resistance (Fig. 3a). Fig. 3b and Fig. S14-S16 illustrate the experimental evaluation of dye removal efficiency for the six dye molecules dissolved in methanol, thereby confirming the rejection mechanism in Fig. 3a. Specifically, the rejection efficiencies for positively charged MB, Su I, and RG are 99.8%, 99.6%, and 98.2%, respectively, while the rejection efficiency for the neutrally charged BRh is 93.8%. In contrast, negatively charged MO and OG exhibit low rejection efficiencies of 23.8% and 29.7%, respectively. Fig. S17 illustrates the methanol permeance measured during filtration process for the different dyes. Fig. 3c compares the performance of the GO@methanol membrane to previous

studies on methanol permeance and MB rejection rates (**Table S3**). GO@methanol membrane achieves the highest methanol solvent permeance of $14,905 \pm 194$ L m⁻² bar⁻¹ h⁻¹, concurrently ensuring high MB dve rejection of 99.8%.

In addition, seven 20-minute interval tests conducted over a twohour period assess the stability of GO@methanol membrane in terms of MB rejection and the permeance (Fig. 3d). GO@methanol membrane maintains a high MB rejection rate of over 99.8%, and the permeance of the MB-methanol mixture remains virtually unchanged over a 120-min period, demonstrating exceptional stability and stable methanol permeance (Fig. S18). Fig. S19 verifies the long-term integrity and stability of GO@methanol membrane immersed in 250 µmol/L methylene blue



Fig. 4. (a) Relationship between assembly time and the thickness of GO@methanol macro-materials, insets are digital photographs of GO@methanol macro-materials with thicknesses of 250 nm, 64 μm, 5 mm and 1.3 cm, respectively. (b) Digital photographs of ultra-light RGO@methanol aerogel supported by petals. (c) Current response curves caused by loading and unloading diverse weights on RGO@methanol pressure sensor. (d) Cycle stability of RGO@methanol pressure sensor within 10,000 s. (e) Schematic diagram of RGO@methanol sensor under vocal movement in the throat. (f-h) Current response curves of RGO@methanol sensor when speaking different words, including "work", "friend", and "green", respectively. (i) EMI SE and (j) shielding mechanism diagram of RGO@methanol aerogel and the reported EMI shielding materials.

methanol solution for 24 hours. The response of the GO@methanol membranes at pH = 5 and pH = 9, as depicted in **Fig. S20**, demonstrated a methylene blue rejection rate of 91.5% at pH = 5.

3.4. GO@methanol macro-materials and their performance for pressure sensors and EMI shielding

As vacuum filtration requires 68.5 h to prepare a GO@water membrane with a thickness of approximately 25 µm, it is not feasible to fabricate aerogels with millimeter or centimeter-scale thicknesses. The rapidly assembled GO@methanol membrane may be employed in the development of pressure sensors. The precise modulation of the GO suspension concentration and solvent volumes facilitates the fabrication of GO gels with varying thicknesses [43]. Through this approach, GO@methanol hydrogels with distinct cake-like morphologies were successfully engineered, demonstrating thicknesses ranging from 250 nm to 1.3 cm, as depicted in Fig. 4a. Concurrently, GO@methanol membranes with diameters ranging from 2.7 cm to 9 cm can be fabricated, as shown in Fig. S21. Fig. S22 demonstrations show that GO @ methanol membranes prepared with different radii show almost no changes. This demonstrates the versatile applicability, scalability, and superior processability of the GO@methanol membrane. After a sequential process comprising water exchange, freeze-drying, and reduction, the resulting GO@methanol gels can be efficiently transformed into reduced GO (RGO)@methanol aerogels. In comparison to the severe conditions, such as elevated temperature and pressure, required for aerogel preparation via the hydrothermal method, the proposed method offers advantage. The lightweight of the RGO@methanol aerogel (1.75 g) are clearly demonstrated by its capacity to be supported on petals, as illustrated in Fig. 4b. To assess its performance as a pressure sensor, a small segment of the RGO@methanol aerogel was positioned between two flag-shaped copper strips to construct the sensor (Fig. S23).

The pressure sensor was subsequently subjected to incremental loads (5 g, 10 g, 20 g, and 50 g), yielding distinct graphs depicting current fluctuations in response to the applied pressure, thereby demonstrating its responsive behavior under varying stress conditions (Fig. 4c). Fig. S24 assesses the sensitivity of the RGO@methanol pressure sensor, and Fig. S25 and Fig. S26 illustrate the pressure sensing mechanism of RGO@methanol pressure sensor [44-46]. The RGO@methanol pressure sensor demonstrates robust cyclic stability across multiple tests conducted over 10,000 s, as illustrated in Fig. 4d. As depicted in Fig. 4e, the RGO@methanol pressure sensor is further utilized for the detection of a crucial signal that necessitates precise monitoring, namely the vocalization process. When positioned on the throat, the RGO@methanol pressure sensor generates a consistent and distinctive pattern that corresponds to the vocalizations of the individual being tested. As illustrated in Fig. 4f-4h, various words provoke distinct and recognizable patterns. This outcome can be attributed to the sensitivity of the RGO@methanol pressure sensor to minute vibrations emanating from the throat and surrounding skin. The RGO@methanol pressure sensor exhibits remarkable capabilities in detecting vocalizations via electromechanical techniques, thereby presenting potential applications in supporting individuals with voice disorders. Moreover, RGO@methanol aerogel-based electromagnetic interference (EMI) shielding blocks exhibit superior performance, demonstrating a high EMI shielding effectiveness (SE) of 83.69 dB and a high SE/density value of 593.55 dB g⁻¹ cm³ compared with reported EMI shielding materials (Fig. 4i–4k, Table S4). In Fig. S27, the RGO@methanol aerogel was applied to material A for electromagnetic shielding, with the R coefficient measured. The T coefficient was also analyzed, based on the relation A +R + T = 1. The A, R and T coefficients were determined to be 0.065, 0.934, and 7.41 \times $10^{-7},$ respectively. The R value significantly exceeds the A value, suggesting that the predominant shielding mechanism is reflection. This can be attributed to the impedance mismatch between the material and free space, which results in most of the incident microwaves being reflected prior to their absorption by the aerogel. Consequently, the reflection of incident electromagnetic waves by RGO@methanol aerogel primarily permits the absorption of smaller electromagnetic waves. Furthermore, the internal multilayer structure of RGO@methanol aerogel induces multiple internal reflections as electromagnetic waves traverse the material. These multiple reflections and scattering significantly enhance the energy transfer of electromagnetic waves, converting the energy into microcurrent-induced heat loss, thereby effectively improving absorption and underscoring their substantial potential for application in lightweight EMI shielding [47,48]. The conductivity properties of the reduced RGO@methanol membrane, RGO@water membrane, and RGO@methanol aerogel are presented in **Fig. S28**.

4. Conclusions

In summary, a novel approach is proposed to enhance solvent channels by constructing a bouffant structure utilizing low-surfacetension solvents, thereby enabling ultra-rapid macroscopic assembly of graphene oxide (GO) membranes in just 10 s, which is 24,660 times faster than that of conventional water-based assembly (over 2.85 days). The accelerated assembly process allows for precise control over the thickness of GO membranes, which can range from the nanometer scale to the centimeter scale, thus effectively addressing the inherent trade-off between assembly rate and scalability of membrane thickness. These versatile GO macroscopic materials demonstrate considerable potential across various applications, including organic solvent nanofiltration at smaller thicknesses, as well as electromagnetic interference (EMI) shielding and pressure sensors at larger thicknesses. The GO membrane employed for dye filtration exhibits a high permeance of 14,905 \pm 194 L $m^{-2} h^{-1} bar^{-1}$ and exceptional selectivity (>99%) of methylene blue and Sudan I. Moreover, RGO@methanol pressure sensors generate distinct electrical signals in response to different vocalizations. RGO aerogelbased EMI shielding blocks also exhibit superior performance, with a high EMI shielding effectiveness (SE) of 83.69 dB and a significant SE/ density value of 593.55 dB g⁻¹ cm³. Our strategies align with recent developments in programmable and adaptive membrane materials.

Data availability

The data supporting this article have been included as part of the Supplementary Information.

CRediT authorship contribution statement

Long Zhang: Writing – original draft, Formal analysis, Conceptualization. Pengfei Chen: Writing – review & editing, Supervision. Zhe Wang: Writing – review & editing, Methodology, Data curation. Hao Feng: Visualization, Investigation. Peng Li: Visualization, Formal analysis. Wei Qian: Methodology, Formal analysis. Huaqiang Fu: Writing – review & editing. Cheng Chen: Writing – review & editing, Methodology. Zuhao Shi: Software, Project administration. Yong Lv: Writing – review & editing. Shuxin Li: Writing – review & editing. Daping He: Validation, Supervision, Funding acquisition.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.surfin.2025.106634.

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