

# Exfoliation of Metal-Organic Framework Nanosheets Using Surface Acoustic Waves

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## Abstract

Two-dimensional (2D) metal-organic framework (MOF) nanosheets have recently received extensive attention due to their ultra-thin thickness, large specific surface area, chemical and functional designability. In this study, an unconventional method using surface acoustic wave (SAW) technology is proposed to exfoliate large quantities and uniform layers of 2D MOF-Zn<sub>2</sub>(bim)<sub>4</sub> nanosheets in a microfluidic system. We successfully demonstrated that the thickness of 2D MOF is effectively and accurately controlled by optimizing the SAW parameters. The mechanisms for the efficient exfoliation of 2D MOF nanosheets is attributed to both the electric and acoustic fields generated by the SAWs in the liquid. The electric field ionizes the methanol to produce H<sup>+</sup> ions, which intercalate Zn<sub>2</sub>(bim)<sub>4</sub> sheets and weaken the interlayer bonding, and the strong shear force generated by SAWs separates the MOF sheets. A yield of 66% for monolayer MOFs with a maximum size of 3.5 μm is achieved under the combined effect of electric and acoustic fields. This fast, low-energy exfoliation platform has the potential to provide a simple and scalable microfluidic exfoliation method for production of large-area and quantities of 2D MOFs.

**Keywords:** metal-organic framework (MOF- Zn<sub>2</sub>(bim)<sub>4</sub>) nanosheets, surface acoustic wave(SAW), electric field

## 1. INTRODUCTION

Compared with three-dimensional (3D) bulk nanomaterials, two-dimensional (2D) nanosheets, such as those based on graphene[1, 2], black phosphorus[3], transition metal dichalcogenide[4], transition metal carbides and nitrides (MXenes)[5], and metal organic frameworks (MOF)[6], have many advantages due to their intrinsic ultrathin structures, excellent mechanical flexibility, good optical transparency, large specific surface area, and abundant catalytic active sites[7]. Among them, 2D MOF nanosheets with their excellent mechanical flexibility[8], adjustable pore size, and chemical and functional designability[9], have shown great potentials for applications in sensing[10, 11], electronics[12, 13], energy[14], gas separation[15], and biocatalysts[16].

The commonly used preparation methods for 2D MOF nanosheets are mainly divided into two categories, i.e., bottom-up and top-down. Bottom-up approaches such as interfacial synthesis[17, 18], surfactant-assisted synthesis[19-21], three-layer modulated synthesis[22], directly achieve the synthesis of 2D MOF nanosheets through metal-containing organic linkers. However, they have disadvantages. For examples, the obtained nanosheets have a thick layer and the surfactant often strongly adheres to the nanosheet surfaces[21, 22]. These defects hinder the exposure of more active sites, and cause major problems for the application of 2D MOFs. The top-down methods, including ultrasonication[23, 24], ball milling[25] and shear mixing[26], are effective in obtaining high yield suspensions of 2D MOF flakes[27]. They mostly rely on the external forces to agitate fluid with high speeds, thus resulting in strong shear forces in the fluid to exfoliate the MOFs. The processing time is varied from ten of minutes to a few hours[24, 26, 28, 29]. However, the thickness distribution is often not uniform, ranging from a few nanometers to hundreds of nanometers[24, 29]. They also have drawbacks including high energy consumption, inability to obtain high yields of monolayer nanomaterials, and difficulty in production of large-scale single layer sheets. Therefore, there is an urgent need to find other types of simple but efficient methods to obtain nanosheets with a monolayer thickness in a large

1 scale.

2 Surface acoustic waves (SAWs) are electromechanical waves with nanometers or sub-nanometer  
3 amplitudes propagating along the surface of a piezoelectric substrate, accompanied by changes in the electric  
4 field along its propagation path. The wave energy is concentrated within one wavelength depth of the device  
5 surface, and the wave energy is dissipated into the liquid, which forms an efficient method for driving liquids  
6 at the microscale level [30, 31]. The large shear force generated in the liquid can effectively exfoliate the bulk  
7 materials into 2D nanosheets[5, 32, 33]. SAWs have many advantages over the conventional ultrasonic waves,  
8 including simple operation, low power consumption, and easy control of acoustic energy[34]. They have  
9 advantages to control the thickness of the exfoliated 2D nanosheets due to its combination of strong acoustic  
10 and electric fields within the liquid. For example, Mohiuddin et. al [32] reported that the electric field  
11 generated by the propagating SAWs plays a dominant role in exfoliating MoS<sub>2</sub> for ultrathin layers with a large  
12 dimensional size. Yang et. al[35, 36] reported that electrochemistry has a significant impact on exfoliation of  
13 2D MOF. However, it is not fully understood how the different and separate effects of electric field and  
14 acoustic field generated by the SAWs within the liquid on the exfoliation principles of 2D materials.

15 In this paper, we use the SAW exfoliation method to prepare MOF-Zn<sub>2</sub>(bim)<sub>4</sub> (bim = benzimidazole)  
16 nanosheets, from bulk Zn<sub>2</sub>(bim)<sub>4</sub> materials. The obtained 2D MOF structure is highly stable and  
17 microcrystalline after exfoliation[37, 38]. 2D Zn<sub>2</sub>(bim)<sub>4</sub> nanosheets have a variety of applications, such as  
18 membranes for separation and purification of hydrogen gas from CO<sub>2</sub> [39], purification of water[37], and laser  
19 desorption/ionization of small molecules[28]. We find that the thickness of the obtained nanosheets can be  
20 easily controlled by regulating the SAW durations and we obtain single layer nanosheets with sizes up to 3.5  
21 μm. Most importantly, we verify that the accompanying electric field generated by the SAW effectively  
22 facilitate the exfoliation process of 2D Zn<sub>2</sub>(bim)<sub>4</sub>. In combination of joint influences of acoustic wave and  
23 electric field generated by SAWs, large-area, uniformly distributed ultrathin 2D Zn<sub>2</sub>(bim)<sub>4</sub> nanosheets have

1 been obtained.

## 2 3 **2. EXPERIMENT SECTION**

### 4 2.1 Fabrication of SAW Device

5 A 4-inch single crystalline LiNbO<sub>3</sub> (LN, 128° Y-rotated X-cut, 1 mm thick) wafer was used as the  
6 substrate. Interdigital transducers (IDTs, 20 pairs) with their reflectors were patterned on the LN wafer, with  
7 a 20 nm chromium adhesive layer and a 80 nm gold layer[40]. The IDTs have a wavelength of 400 μm  
8 (corresponding to the resonance frequency of 9.7 MHz) and its reflector of 100 μm width. The resonant  
9 frequency of the device was characterized using a vector network analyzer (N5232A). A signal generator  
10 (AFG3022, USA) and an amplifier (TREK MODEL 2100HF) were used to apply RF signals to the SAW IDTs  
11 at its resonance frequency of ~9.7 MHz for the acoustofluidic tests.

### 12 2.2 Preparation of PDMS liquid reaction reservoir

13 PDMS curing agent was mixed with the prepolymer at a mass ratio of 1:10. The mixture was  
14 ultrasonically stirred for 5 min, and then the air bubbles were removed by keeping it in a vacuum chamber.  
15 The prepared PDMS solution was poured into a plastic Petri dish to form a layer of 2 mm thick and cured in  
16 an oven. The curing temperature was 80°C for 1 h. After curing, the PDMS chamber was prepared by  
17 punching a hole with a diameter of 9 mm for microchamber connection. The PDMS chamber and the LN  
18 wafer with IDTs were then placed inside a plasma chamber and treated with plasma. Then the treated PDMS  
19 layer was pressed onto the LN layer for bonding, and the bonded device was placed in an oven at 110°C for  
20 1 h, before cooling down to room temperature[41]. The fabricated SAW device for exfoliation is shown in  
21 Figure S4.

### 22 2.3 Preparation of bulk Zn<sub>2</sub>(bim)<sub>4</sub>

23 Bulk Zn<sub>2</sub>(bim)<sub>4</sub> precursors were prepared based on the hydrothermal transformation of ZIF-7

1 nanoparticles[25]. Firstly, 0.605 g of zinc nitrate hydrate ( $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ , 99.99%) and 1.539 g of  
2 benzimidazole (bim, >98%) were added in 200 mL of N,N-dimethylformamide (DMF, AR). After stirring for  
3 1 h, the mixed solution was left for 72 h at room temperature. The formed ZIF-7 nanoparticles were collected  
4 by centrifugation and washed with methanol. The collected sediment was dried at 50 °C for 12 h, followed  
5 by drying in a vacuum oven at 120 °C for 48 h. Next, 0.34 g dried ZIF-7 nanoparticles were dispersed in  
6 deionized (DI) water up to a concentration of 0.5 wt % and then heated at 100 °C for 24 h. Finally, the  
7  $\text{Zn}_2(\text{bim})_4$  was obtained by centrifugation and washed with DI water and methanol. The collected sediment  
8 was dried at 50 °C overnight.

#### 9 2.4 Nanosheet preparation

10  $\text{Zn}_2(\text{bim})_4$  powder of 2.5 mg was added into 10 mL MeOH/n-propanol mixture (V: V = 1: 1). In the  
11 exfoliation process, the above liquid mixture with a volume of 120  $\mu\text{L}$  was put into the PDMS reservoir. The  
12 operating frequency of the SAWs device was 9.7 MHz and the applied voltage was 15 V. The SAW agitation  
13 durations varied from 1, 10, 20, 60 and 90 min. After the exfoliation, the liquid in the PDMS chamber was  
14 transferred into a 1.5 ml centrifuge tube and diluted to 1 ml solution with a mixture of MeOH/n-propanol. The  
15 supernatant was collected for characterization after it was centrifugated at 9000 rpm for 25 min. The collected  
16 supernatant was dropped into the PDMS chamber on the SAW device and dried in a vacuum oven until the  
17 supernatant was changed into white  $\text{Zn}_2(\text{bim})_4$  nanosheet powder.

#### 18 2.5 Characterization

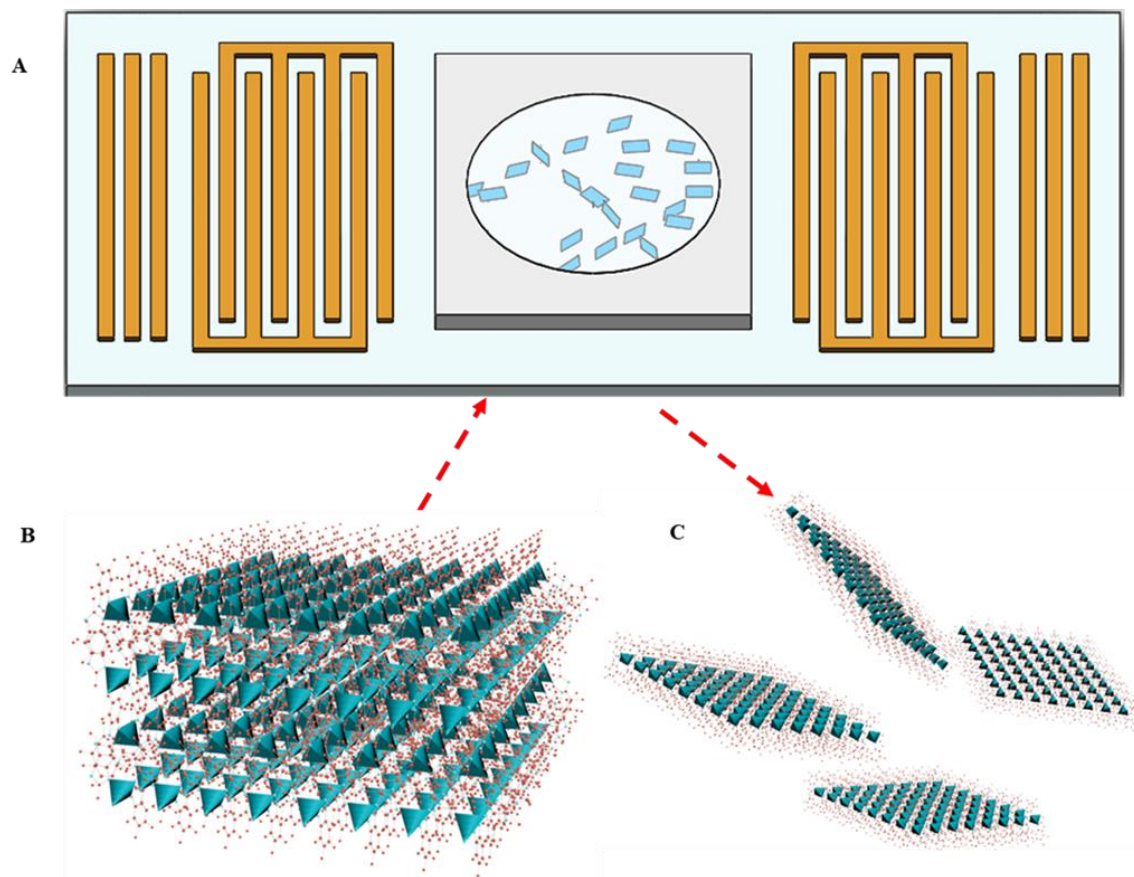
19 Crystalline structures of both the prepared  $\text{Zn}_2(\text{bim})_4$  bulk and  $\text{Zn}_2(\text{bim})_4$  nanosheets were characterized  
20 using X-ray diffraction (XRD, D8 ADVANCE A25, Germany). Morphology of  $\text{Zn}_2(\text{bim})_4$  bulk was  
21 characterized using a scanning electron microscope (SEM, SU8000, Japan).  $\text{Zn}_2(\text{bim})_4$  nanosheet suspensions  
22 were added onto a copper-supported ultrathin carbon grid (200 mesh, Chukyo Kiyoko Co.), for characterization  
23 using a transmission electron microscope (TEM, JEOL JEM-F200) and a high-resolution TEM (HRTEM,

1 JEOL JEM-2100Plus) with the attached energy dispersive X-ray spectroscopy (EDX). The  $Zn_2(bim)_4$   
2 nanosheet suspension was dropped onto the silicon substrate (1 cm×1 cm), and then dried in a vacuum oven  
3 to characterize the surface topography and thickness profile using an atomic force microscope (AFM, Bruker  
4 Dimension Icon, INNOVA). At least 50 particles per experimental condition were tested during the AFM  
5 analysis and the average readings were obtained.

6

### 7 3. RESULTS AND DISCUSSION

8 Figure 1 shows the exfoliation device, including a lithium niobate ( $LiNbO_3$ , LN) wafer and a  
9 polydimethylsiloxane (PDMS) chamber. The illustrations of  $Zn_2(bim)_4$  structure before and after the  
10 exfoliation processes are represented in Figures 1B and 1C, respectively.



11

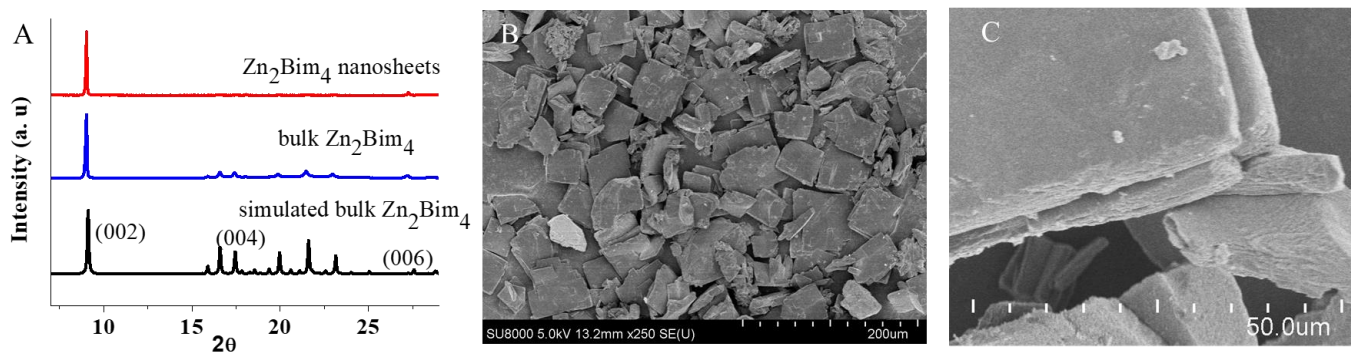
12 Figure 1. Illustrations of SAW exfoliation device and  $Zn_2(bim)_4$  structures. (A) Illustration of the SAW exfoliation device

13 constituting an LN chip with a PDMS reservoir bonded onto it. (B) Illustration of bulk  $Zn_2(bim)_4$  structure. (C) Illustration

14 of monolayer  $Zn_2(bim)_4$  nanosheets structure.

1

2 Figure 2A shows the XRD pattern of bulk  $Zn_2(bim)_4$ , which is matched with the reference of (CCDC-  
3 675375)[25], proving that the  $Zn_2(bim)_4$  was successfully synthesized. Figure 2B shows an SEM image of  
4 the bulk  $Zn_2(bim)_4$ , showing numerous block structures. Figure 2C shows an SEM image of the typical  
5 layered structure of the  $Zn_2(bim)_4$ .

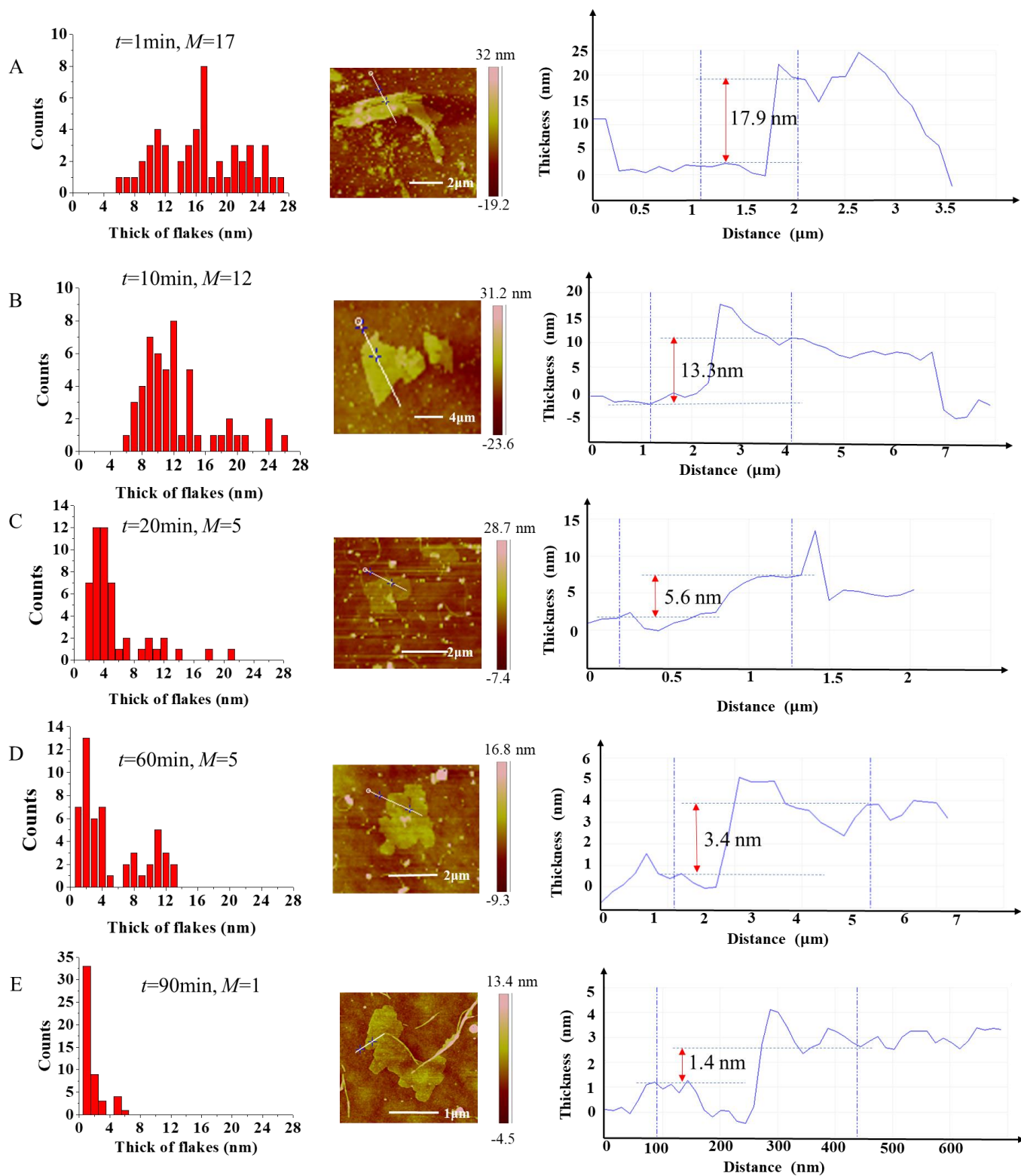


7 Figure 2. Characterization of bulk  $Zn_2(bim)_4$ . (A) The XRD patterns of bulk  $Zn_2(bim)_4$  and  $Zn_2(bim)_4$  nanosheets. (B) The  
8 SEM image of the bulk  $Zn_2(bim)_4$  materials. (C) The typical layered structure morphology of  $Zn_2(bim)_4$ .

9

10 The bulk  $Zn_2(bim)_4$  material was exfoliated by the SAWs device as shown in Figure S5. Figure 3 shows  
11 the AFM images of  $Zn_2(bim)_4$  nanosheets obtained from samples with SAW exposure durations from 1 to 90  
12 min, along with their thickness distribution histograms. Figure 3A shows a large thickness distribution in the  
13 range 6-27 nm ( $M=17$  nm,  $M$  represents the median thickness of 50 nanosheets) for the sample exposed to  
14 SAWs for 1 min. With increasing the exposure time, the thicknesses of nanoflakes gradually decreased as  
15 shown in Figures 3A to 3E. At an exposure time of 20 min (Figure 3C), the thickness of nanosheets was 5 nm  
16 (e.g.,  $\sim 5$  layers with a single layer thickness of  $\sim 0.98$ [25]). The SAW exfoliated sample for 20 min was  
17 analyzed by XRD, and the result is shown in Figure 2. It matches well with that of the bulk  $Zn_2(bim)_4$ , which  
18 demonstrates that the exfoliation process did not destroy the crystalline structure of  $Zn_2(bim)_4$ . When the SAW  
19 exposure time increased to 90 min, almost 90% of the nanosheets samples was less than 3-layer thick as shown

1 in Figure 3E. The estimated yield of monolayers obtained is 66%, which is the percentage of monolayer  
 2 nanosheets obtained from 50 nanosheets.



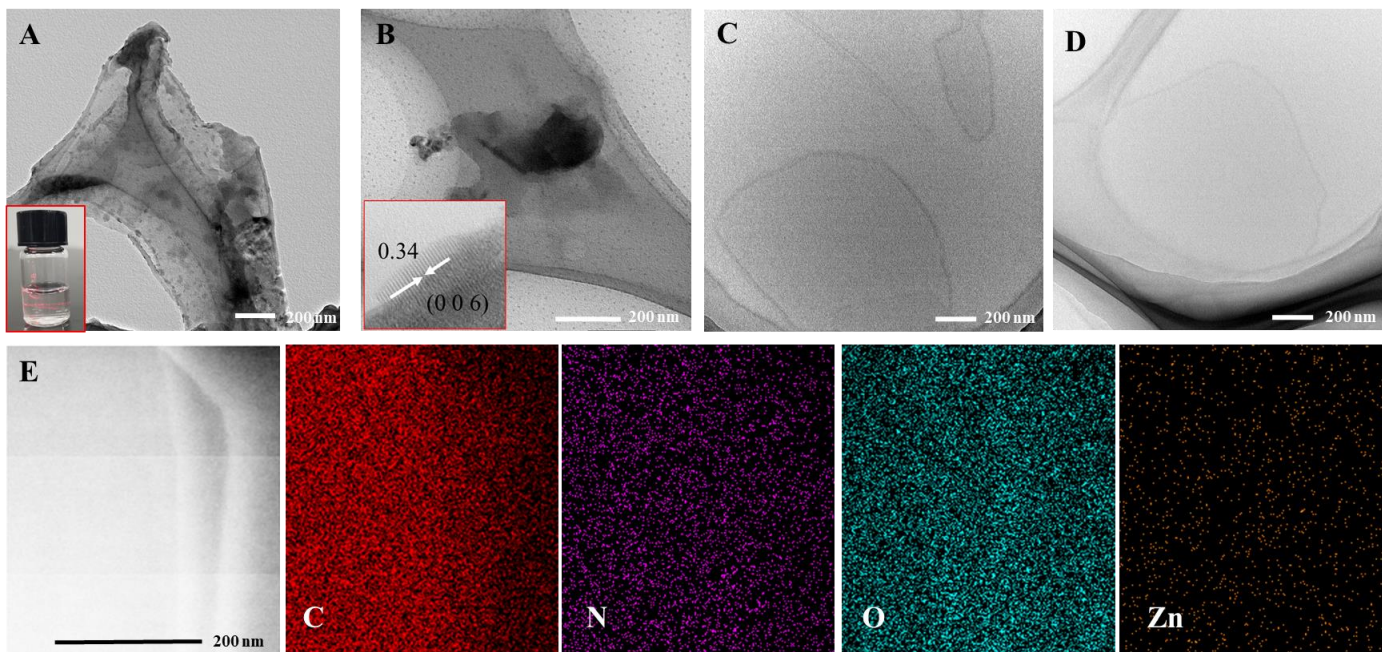
3  
 4 Figure 3. Thickness distribution of  $\text{Zn}_2(\text{bim})_4$  nanoflakes obtained from the AFM measurements. Thickness distribution  
 5 histograms of the exfoliated  $\text{Zn}_2(\text{bim})_4$  nanoflakes, along with typical images and height profile at different SAW exposure



1 durations of (A) 1min, (B) 10min, (C) 20min, (D) 60min and (E) 90min.  $t$  represents the exposure time and  $M$  represents the  
2 median thickness of 50 nanosheets.

3

4 Figure 4 shows TEM and HRTEM images of  $\text{Zn}_2(\text{bim})_4$  nanosheets exposed to SAWs for 20 min and 90  
5 min, respectively, along with the energy dispersive X-ray (EDX) mapping patterns of elements. The Tyndall  
6 effect of  $\text{Zn}_2(\text{bim})_4$  nanosheet solution (the inset in Figure 4A) was observed which confirms that they are  
7 colloidal materials[42]. TEM images of the obtained  $\text{Zn}_2(\text{bim})_4$  nanosheets (Figure 4A and 4B) show wrinkle  
8 patterns, which indicate the flexibility of the obtained nanosheets[43]. The HRTEM (the inset in Figure 4B)  
9 image shows that the fringe spacing of the  $\text{Zn}_2(\text{bim})_4$  nanosheets is  $\sim 0.34$  nm, corresponding to the (006)  
10 lattice plane of the  $\text{Zn}_2(\text{bim})_4$  with a lattice constant of  $\sim 0.32$  nm[25]. Figures 4C and 4D show that the  
11 monolayer materials have been obtained when the exposure time is 90 min. The EDX elemental mapping  
12 images in Figure 4E show that C, N, O and Zn are distributed uniformly inside the nanosheets[26, 44, 45].



13

14 Figure 4. TEM image of a piece of  $\text{Zn}_2(\text{bim})_4$  nanosheets. TEM images of the  $\text{Zn}_2(\text{bim})_4$  nanoflakes for different SAW  
15 exposure durations: (A) 20 min samples with tyndall effect image of  $\text{Zn}_2(\text{bim})_4$  nanosheets, (B) 20 min samples with a HRTEM  
16 image, (C) and (D) 90 min samples. (E) The EDX mapping image of  $\text{Zn}_2(\text{bim})_4$  nanosheet.

17

1 To investigate the effect of electric field generated by the SAWs on exfoliation of the MOFs, we have  
2 made two additional microchips for exfoliation, which are different from the SAW chip (as shown in Figure  
3 S7 (A)).

4 The first one (Figure S7 (B)) only produces an acoustic wave field, by depositing a metal (20 nm thick  
5 Cr adhesive and 80 nm thick Au) layer between the PDMS chamber and the LN substrate. The metal layer  
6 between the LN substrate and PDMS can shield the electric field after the voltage is applied. The second one  
7 (Figure S7 (C)) can only generate an electric field with the IDTs patterned on glass substrate (without any  
8 piezoelectric material) and the PDMS chamber pressed onto the IDTs position. The obtained current-voltage  
9 ( $I$ - $V$ ) curves are shown in Figure 5A, which reveals the currents in methanol/n-propanol mixture on three  
10 different chips. The electrolytic current becomes much larger when the SAW driving voltage is increased from  
11 0 V to 15 V. This is mainly due to generation of more  $H^+$  ions from the ionization of methanol, when the  
12 platform is applied with ultra-high voltages [46]. On the LN substrate chip with a metal layer, the  $I$ - $V$  curve  
13 (Figure 5A) under the voltage of 15V is similar to that without using the SAW (SAW, 0V), which means that  
14 no electric field was generated in the liquid with the addition of this metal layer. By applying an external  
15 voltage to the IDTs which on the glass substrate, the current generated in the solution is same with that under  
16 the agitation of the SAWs (with a voltage of 15 V). The obtained AFM images of the exfoliated MOFs after  
17 90 minutes from these chips are shown in Figures 5B-5D. From these figures, the largest dimension size of  
18 the nanosheet is obtained under the combined effect of acoustic and electric fields, and then the smallest  
19 transverse size of the nanosheet is obtained under the electric field only.

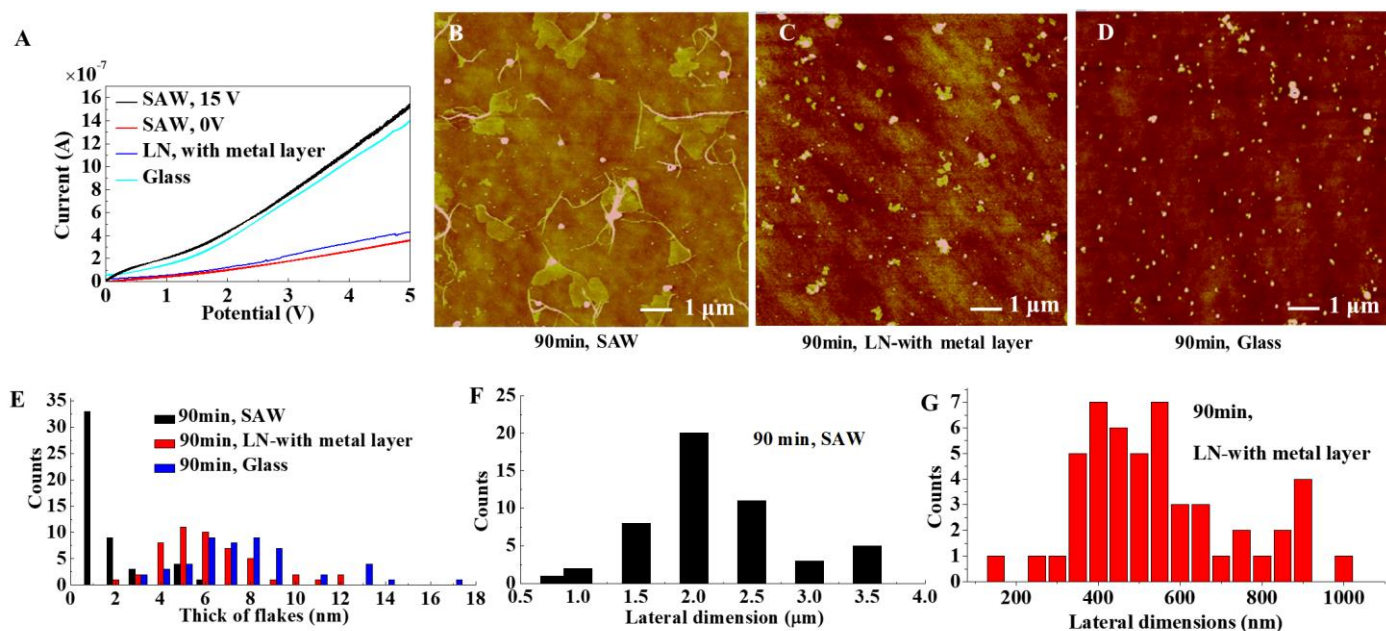


Figure 5. Comparison of nanosheets data with different chips. (A)  $I-V$  curves for MeOH/n-propanol mixture liquid measured using a two-wire probe station. The current flow was monitored through a voltage sweep from 0 to 5 V, under SAW excitation, LN substrate with metal layer chip and glass substrate chip. The AFM pictures of (B) SAW chip, (C) LN substrate with metal layer chip and (D) glass substrate chip. (E) Thickness distribution histograms of  $Zn_2(bim)_4$  flakes (90 min) under SAW chip (black), LN substrate with metal layer chip (red), and glass substrate chip (blue). (F) Nanoflakes lateral dimension histograms under SAW chip for 90min. (G) Nanoflakes lateral dimension histograms under LN substrate with metal layer chip for 90min.

Under the effect of acoustic field only (e.g., the LN chip with a metal layer), the average thickness of the nanosheet obtained is  $\sim 6$  nm (with the red color in Figure 5E). They are much thicker than the nanosheets (in black color) obtained under the agitation of both acoustic and electrical fields (Figure 5E). Figure 5F shows the lateral dimension of the nanosheets using the SAW agitation, with dimensions up to 3.5  $\mu\text{m}$  and an average size of 2  $\mu\text{m}$ . In contrast, on the LN substrate chip with only a metal layer (e.g., only electrical field effect), the lateral dimensions of the nanosheets are much smaller, with an average size of 540 nm as shown in Figure 5G.

The above results clearly show that the electric field plays an import role in the exfoliation process and

1 can be applied to achieve 2D nanosheets with large cross-sectional area and ultrathin thickness. Under the  
2 action of electric field, the solution is ionized, and the insertion of  $H^+$  ions into the layers of 3D MOF  
3 significantly expands the distance between the layers and weakens the interlayer bonding strength. This will  
4 promote the separation of different layers and also the subsequent release of the 2D flakes into the solution[35,  
5 36, 47]. Whereas on the glass substrate with only the electric effect (without acoustic field), the average  
6 thickness of  $Zn_2(bim)_4$  nanosheets is  $\sim 8$  nm (as shown in blue color of Figure 5E). Its lateral dimension is less  
7 than 200 nm which can be seen from the AFM picture shown in Figure 5D. The above results confirm that  
8 the applied electric field can promote the fast exfoliation of the bulk MOF, but the size of the obtained  
9 nanosheets is small, without any single layer nanosheet generated.

10 In addition, to further understand the mechanism that underpins the acoustic field and electricity field  
11 exfoliation process, we have applied two different methods for exfoliation of the MOF nanosheets ( $Zn_2(bim)_4$ ).  
12 The first method we used to exfoliate the  $Zn_2(bim)_4$  is under the acoustic fields for 90 min (using the LN chip  
13 with a metal layer), and then applied with the electric field (using the glass chip) for another 90 min. A  
14 schematic diagram of this method is shown in Figure 6A. The second method is to apply the electric field  
15 (using the glass chip) for 90 min, and then apply the acoustic field using the LN chip with a metal layer) for  
16 another 90 min, as shown in Figure 6B.

17 Figures 6C to 6F show the AFM images and thickness distribution histograms of the samples obtained  
18 from these two methods. For the sample prepared using the first method, the average thickness of the  
19 nanosheets obtained is 8 nm and the lateral size is less than 200 nm, as shown in Figures 6D. After 90 min of  
20 acoustic field action, followed by the electric field exfoliating for 90 min, the nanosheets (Figure 5C) are  
21 mostly fractured laterally due to the intercalation of  $H^+$  ions. The thickness of these 2D MOFs hardly thins  
22 further. Whereas for the sample prepared using the second method, we obtained 6 nm thick nanosheets with  
23 lateral dimension less than 200 nm as shown in Figure 6F. This is because the size of the nanosheets (Figure

5D) obtained by applying only the electric field is so small that the acoustic waves cannot continue to peel them effectively. Clearly, the results of nanosheets obtained from these two methods are similar, while they are both much worse than those achieved by using a standard SAW device. The results shown in Figure 3E clearly demonstrate that the combination of acoustic and electric fields can effectively promote the formation of single-layer MOFs.

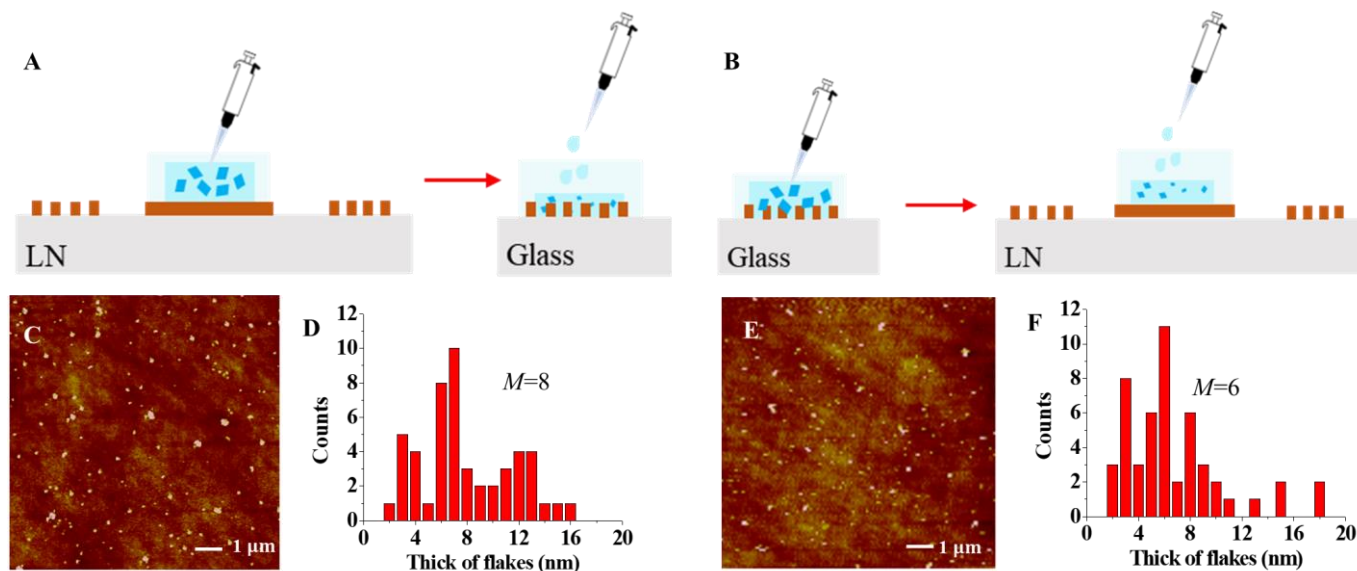


Figure 6. Schematic illustration of two different methods, and thickness images obtained by AFM and thickness distribution histograms of the exfoliated  $Zn_2(bim)_4$  nanoflakes. (A) Schematic illustration of method 1, (C) AFM images and (D) thickness distribution histograms of the exfoliated  $Zn_2(bim)_4$  nanoflakes using the first method (i.e., acoustic field for 90 min, then electric field for 90 min). (B) Schematic illustration of method 2, (E) AFM images and (F) thickness distribution histograms of the exfoliated  $Zn_2(bim)_4$  nanoflakes using the second method (i.e., electric field 90min, then acoustic field for 90 min)  $M$  represents the median thickness.

## 5. CONCLUSIONS

A SAW device on a LV substrate is used in this paper to apply both acoustic field and electric field to effectively exfoliate bulk MOF- $Zn_2(bim)_4$  into monolayer-enriched 2D  $Zn_2(bim)_4$ . The thickness of the  $Zn_2(bim)_4$  can be modulated by changing the exposure time of SAW. When the SAW exposure time is 20 minutes, the average thickness can reach to 5 nm. With the continuous increased of the exposure time to 90

1 min, ~66% of monolayers  $Zn_2(bim)_4$  and the maximum lateral dimension up to ~3.5  $\mu\text{m}$  have been achieved.  
2 We have investigated the effects of both acoustic field and electric field effects for exfoliation process. By  
3 shielding the accompanying electric field and using only acoustic waves induced by SAW, the obtained  
4 nanosheets are thicker and have smaller transverse dimensions. When only the electric field or only the  
5 acoustic wave field is applied to exfoliate bulk MOF- $Zn_2(bim)_4$ , nanosheets with an average thickness of 6 or  
6 8 nm and a small transverse dimension are obtained. By combining these two effects of acoustic and electric  
7 fields simultaneously (e.g., using SAWs), much better exfoliation results are obtained with a high percentage  
8 of monolayers and in the micron range dimensional size.

## 9

## 10 **ASSOCIATED CONTENT**

11 Supporting information. Simulation section, photographs of SAW exfoliation device, XRD patterns and  
12 nanosheets data with different chips were included in supporting information.

## 13

## 14 **Notes**

15 The authors declare no competing financial interest.

## 16

## 17 **ACKNOWLEDGMENTS**

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