



A Concise Overview of Ultrasound-Assisted Techniques for the Production of 2D Materials

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Abstract: The production of low-dimensional materials is the key topic of nanoscience. The development of new routes to downsize organic and inorganic materials has focused the attention of a great part of the scientific community that is still debating on the best route to pursue. Among nanostructures, 2D species have been investigated since the discovery of graphene. Nonetheless, the production of 2D materials is very complex, and the discussion on which is the most profitable way is still open. Ultrasound-assisted techniques represent one of the best routes for the production of 2D materials with minimum consumption of energy and best performances. Accordingly, we present a concise and exhaustive discussion about the use of ultrasound-assisted techniques for the production of both organic and inorganic 2D materials, also providing a theoretical overview of the mechanism behind the use of ultrasounds in synthetic material science.

Keywords: 2D materials; ultrasound sonochemistry; fabrication processes

1. Introduction

During the first two decades of the 21st century, increasing environmental concerns have heightened the need for accountability in monitoring, preserving, and improving the quality of water, air, and soil [1-3]. Mankind is currently facing the threat of worldwide ecosystem degradation caused by anthropogenic activities by developing new solutions able to mitigate the impact [4]. Finding and destroying pollutants in this global issue is particularly challenging due to the complexity of the problem. Some of these pollutants are called recalcitrant species, since they are difficult to eliminate, while others are newly introduced to ecosystems and are referred to as emerging pollutants [5]. Nevertheless, the scientific community has invested considerable effort into the study and optimization of advanced materials to contribute to solving this issue. Among the various proposals, nanotechnologies have been recognized as the most effective solution [6], building upon neglected dimensions studied in the early 20th century [7]. In particular, 2D materials have played a significant role due to their exotic properties of electronic conduction and transport, optical properties, and physiochemical characteristics [8]. Nonetheless, the production of 2D materials faces notable challenges related to control and scalability. The superior performances of nanostructured and nanosized materials are offset by several issues, including the complexity of techniques used [9,10] and the massive use of chemicals [11,12]. Accordingly, new approaches have been explored to achieve a balance between process efficiency and sustainability [13]. Ultrasound-assisted techniques (UATs) have garnered



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Copyright: © 2024 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/). interest as a green and efficient method for producing 2D materials [14]. UATs have been applied in various fields such as catalysis [15,16], nanoparticle preparations [17], filler dispersion into polymeric matrices [18–22], and biomedical treatment [23]. The main advantages of the UATs are related to the very effective heat and mass transport phenomena induced by the cavitation effect, which boosts the kinetics of several processes [24,25]. In this review, our focus is on the role of UATs in the production of 2D materials, categorizing them into two main groups: graphene-related materials and inorganic materials. We provide an exhaustive description of UAT theory and engage in a comprehensive discussion on recent achievements. Furthermore, we critically compare UATs with other processes including physical and chemical vapor deposition, microwave-assisted synthesis, and mechanochemistry, highlighting their strengths and weaknesses.

2. The Physiochemical Phenomena of UATs

Ultrasound is an acoustic wave that falls within a frequency range of 20 kHz and 10 MHz. The upper-frequency limit is typically set at 5 MHz for gases and 500 MHz for liquids and solids [26], with operative powers ranging from 100 up to 2000 W [27]. Ultrasounds interact with any substance that has elastic properties, transmitting vibrations and inducing continuous movements in the medium.

Ultrasounds exert physical forces that find application in three main types of set-ups: (i) low-power ultrasound with high frequency (1–10 MHz); (ii) medium-power ultrasound (100 kHz–1 MHz), and (iii) high-energy ultrasound with low frequency (20–100 kHz). Low-amplitude waves are commonly used for physical characterization, such as measuring the absorption coefficient of waves in the medium. High-energy waves, on the other hand, are typically employed in ultrasonic cleaning, chemical processes, and dispersion. Chemical species interact with sound waves through indirect physical forces generated by the propagation of energy waves, which creates bubbles in liquids. This phenomenon is known as acoustic cavitation, wherein bubbles form, grow, and violently collapse within a liquid when exposed to sound waves [28–31].

Acoustically induced cavities can be classified into two types: stable and transient. Transient cavities only exist for a short duration, usually less than a single acoustic cycle, rapidly expanding before violently collapsing and often disintegrating into smaller bubbles. On the contrary, stable cavities oscillate around an equilibrium size and can persist for several cycles, although they can also be converted into transient cavities [32]. The collapse of these cavities may emit faint visible light, a phenomenon known as sonoluminescence, which was discovered in 1933 [33].

The study of acoustic cavitation encompasses a wide range of practical processes and serves as a valuable tool for scientific investigations across various sizes and energy scales [34]. Several theories have been proposed to explain how cavitation can influence chemical reactions, including the widely accepted hot-spot theory [35,36], the electrical theory [37], and the plasma theory [38,39].

Figure 1 illustrates the inertial and non-inertial types of cavitation phenomena, where inertial cavitation involves the collapse of bubbles and non-inertial cavitation is characterized by bubble oscillation.



Figure 1. Scheme of the processes of inertial cavitation (**a**) and non-inertial cavitation (**b**) under ultrasound solicitation. Reproduced with permission from Vyas et al. [40].

In an ultrasonic field, the oscillation of molecules in a liquid leads to changes in the distance between them. When a sufficiently large negative pressure is applied to the liquid, the molecules reach a critical distance, causing the liquid to break down and form a cavity. Additionally, bubbles can be generated from pre-existing impurities [26,41].

The size of the bubble is influenced by various factors, including acoustic pressure, ultrasonic power and frequency, and the viscosity of the medium. During the expansion phase, the bubble creates a low-pressure region around it, causing dissolved gases and solvent vapor to migrate into the bubble, resulting in a decrease in bubble thickness. In the contraction phase, the bubble compresses, leading to an increase in the shell thickness. The high internal pressure of the bubble causes mass transport from the bubble to the surrounding liquid. This trend of shell thickness is known as the shell effect.

Crum described the rectification of mass occurring during ultrasound irradiation in a liquid in terms of the area effect and shell effect [42]. The bubble undergoes repeated cycles of expansion and contraction, and its collapse is rapid, providing less surface area for mass transport. As a result, a smaller amount of material diffuses out compared to the amount that diffuses in during the initial phase, known as the area effect. The combination of the shell and area effects effectively explains the expansion and compression phases of bubbles [43].

The subsequent sections will thoroughly discuss the effectiveness of these effects in the synthesis of nanomaterials.

3. UATs for the Production of 2D Materials: Graphene and Related Materials

Graphene is a material composed of a single infinite plane of sp² carbon atoms, which enables a complete delocalization of the π bond system on the graphene plane [44–46]. This unique structure gives rise to impressive electrical conductivity [47], making graphene

applicable in a wide range of fields, such as electronics, energy storage, sensing, drug delivery, etc [48–50].

Despite its remarkable properties, graphene faces several challenges in terms of production and workability [51]. To overcome these challenges, the use of graphene derivatives has become popular as a practical compromise between the desirable properties of graphene itself and the need for more user-friendly working conditions. The primary graphene derivatives utilized are graphene oxide (GO) and reduced graphene oxide (rGO), produced through the exfoliation and oxidation of graphitic precursors, as depicted in Figure 2.



Figure 2. Scheme of (**a**) pristine graphene, GO, rGO, and (**b**) their production. Reprinted from Abu-Nada [52] under CC license 4.0.

Graphene oxide (GO) is a form of oxidized graphene that is modified with oxygencontaining functionalities [53]. These functionalities are distributed on the edges and throughout the core of the material, following the Lerf mode [47]. Reduced graphene oxide (rGO), on the other hand, is a form of GO that has undergone a reduction process, resulting in properties that are closer to those of original graphene flakes as the oxygen content decreases [54].

Alternatively, graphene and related materials can be produced and modified by incorporating organic molecules and inorganic species. This approach allows for the tuning of their solubility and chemical reactivity [55]. There are various routes available for carrying out these treatments, but ultrasound-assisted techniques (UATs) have emerged as one of the most widely used and effective methods, as discussed in the following sections.

3.1. Exfoliation/Oxidation

The synthesis of graphene presents a challenging trade-off between yield and control over the resulting properties of the material [56]. Mechanical exfoliation of graphite without chemical functionalization, for instance, suffers from low production yields, highlighting the need to develop cost-efficient and environmentally friendly methods for synthesizing graphene materials with well-controlled properties. A key objective in graphene synthesis is to obtain nearly pristine graphene with low oxygen content, a reduced number of layers, and a large lateral size. However, exfoliating graphite in liquid electrolytes under ambient

conditions commonly leads to the introduction of oxygen into the carbon lattice of graphene and the fragmentation of exfoliated graphene into smaller pieces.

In addition to its cost-effectiveness and scalability, the shear exfoliation method has been reported to produce high-quality graphene sheets. These sheets exhibit a thickness of up to 1.68 nm and a lateral size ranging from 0.5 to 2 μ m, demonstrating a low defect density. Another method for producing high-quality graphene nanosheets with low defect density in substantial quantities is liquid-phase shear exfoliation under ambient conditions, which utilizes organic solvents such as 1-methyl-2-pyrrolidinone and incorporates urea as a stabilizer [57].

Drawing inspiration from the concept of tape-assisted graphene exfoliation, which is not scalable, two mechanical approaches, namely shear exfoliation and ball milling, have been devised and successfully demonstrated as effective methods for exfoliating layered materials in significant quantities within a single operation [58–60].

As an alternative to mechanical exfoliation methods, the hydrothermal method was initially suggested. Researchers commonly employ two main strategies, namely intercalation (using ions or molecules as intercalants) and chemical reaction, to separate nanosheets from bulk crystals. These strategies are often combined to enhance the efficiency of the process. The hydrothermal process involves treating an aqueous suspension of layered material with chemical and thermal disturbance, providing the necessary energy for the separation of nanosheets from their bulk crystals.

Liquid-phase exfoliation assisted by sonication can be performed using ultrasonication or tip sonication of layered materials in solvents [61]. This technique offers several advantages, including ease of production, preservation of the layered material's structure, and applicability to a wide range of layered materials. However, it also presents some disadvantages, such as difficult scalability to the ton level, difficulties in controlling lateral size and thickness, limited achievable size, and low yields. To address these limitations, Price et al. [62] used a transient cavitation approach in methylnaphthalene to produce graphene oxide (GO) with maximum yields of up to 5.7×10^{-11} kg J⁻¹, even though the production rate reached up to 2.2×10^{-9} kg s⁻¹.

The exfoliation of graphite into graphene is a crucial step in obtaining high-quality graphene. Traditional exfoliation methods often involve the use of organic solvents and surfactants, which can introduce impurities and hinder the scalability of graphene production. In recent years, ultrasound-assisted exfoliation has emerged as a promising technique for obtaining graphene dispersions with improved quality and processability. Soltani et al. [63] reported a fast and cost-effective one-step ultrasonication-assisted treatment for the production of reduced graphene oxide (rGO), reducing the number of layers of each flake down to four with an interlayer spacing of close to 1 nm, without using hydrazine.

Several examples of ultrasound-assisted synthesis of graphene and GO have been reported [64,65] with a particular focus on liquid-phase exfoliation, which remains the most commonly used method for exfoliating 2D materials. Lavagna et al. [66] studied the effect of ultrasonication time on the quality and functionalization degree of GO, showing that the oxidation degree reached a maximum after 60 min, and further increments of processing time did not significantly affect the oxidation of GO. Similarly, Ručigaj et al. [67] correlated oxidation with the amplitude used during the oxidation process, indicating that a value close to 60% induced the highest degree of oxidation.

Furthermore, Sontakke et al. [68] reported the possibility of transforming simple GO into nanoscrolls by tuning ultrasound frequency and power.

3.2. Chemical Tailoring

Since the introduction of this new research area, sonochemistry has had an important impact on chemical synthesis. The study of the reaction and the micro/nano structure formed under the action of ultrasound constitutes a new methodology that often surpasses conventional thermal protocols [69]. The reaction rate improvements have led a significant

number of scientists to apply sonochemistry to the development of various materials, including 2D graphene-based materials [70,71].

These latter materials show interesting properties, such as high surface area, ballistic electrical conductivity [72], and good thermal stability [73], thus making graphene an excellent candidate to host active nanomaterials for various applications. Graphene functionalization affects the architecture of the composites. The final properties of graphenebased material depend on the final structure of graphene or graphene composites [74]. The predictability of the material is complex because structures are affected by lots of parameters, such as frequency, input power, ultrasound exposure time, and solvents [75,76].

Synthesis methods for graphene-based materials can be categorized into atomization synthesis methods, ultrasonic-assisted hydrothermal methods, co-precipitation, and electrodeposition [77]. Ultrasonic atomization is used to produce mist droplets [76] and requires a specialized ultrasonic transducer [78,79]. The flow rate of the gas carrier in the mist is adjusted to control the mist droplets and vapor mixture. Ultrasonic atomization technology can also be employed to produce hollow microspheres without the use of templates or surfactants. For example, in 2016, Yudin et al. proposed the synthesis of nickel oxide nanostructured powders and prepared hollow NiO microspheres using ultrasonic atomization [80].

The ultrasonic-assisted hydrothermal method promotes the dispersion of materials on the carrier surface and introduces active species into the structure to enhance catalytic activity. In 2021, Wang et al. proposed an ultrasound-assisted hydrothermal method to prepare cobalt oxide-incorporated nitrogen-doped graphene (Co_3O_4/N -GO) hybrids [81]. The ultrasonic-assisted co-precipitation method generates nanomaterials with a smaller average particle size and a narrower particle size distribution compared to the traditional co-precipitation method. In 2016, Zhao et al. [82] proposed the ultrasonic-assisted co-precipitation method for ultrathin nanostructured MnO_2 on graphene sheets for electrochemical capacitors. Ultrasonic-assisted electrodeposition is an effective and alternative method for preparing composite coatings with precise compositions and microstructures. Aksoy et al. [83] reported an efficient approach for the production of graphene oxide (GO) by ultrasonic-assisted electrochemical exfoliation. In 2023, Zhan et al. [84] proposed a synthetic method for GO/Cu composites via ultrasonic treatment-assisted electrodeposition.

One of the major applications of graphene-based materials is in the field of electrochemistry, particularly in energy storage/generation devices [85]. In 2015, Raj et al. proposed ultrasound-assisted synthesis at room temperature for Mn_3O_4 nanoparticles anchored on graphene nanosheets without the use of templates or surfactants for supercapacitor applications [86]. They developed a simple synthesis in which both the formation of Mn_3O_4 nanoparticles and the reduction of graphene oxide occurred simultaneously. Similarly, Choudhury et al. [87] developed a high-surface-area Fe_3O_4/rGO nanocomposite via an ultrasound-assisted low-temperature method. Ultrasound with a frequency of 37 kHz was used to disperse GO nanosheets and assist the nucleation of Fe_3O_4 nanoparticles on rGO. They designed a solid-state symmetric supercapacitor with excellent electrochemical properties, as Fe_3O_4 nanoparticles were uniformly grown on the surface of highly porous rGO.

Yang et al. [88] covalently functionalized graphene nanosheets with 3-aminopropyltriethoxysilane. They produced GO nanosheets containing different chemical species such as hydroxyl, epoxide functional groups, carbonyl, and carboxyl groups. Additionally, Quin et al. [89] developed ultrasound-assisted techniques for tailoring GO with folic acid and polyvinylpyrrolidone for the preparation of 2D drug delivery systems.

Nitrogen-rich graphene derivatives represent another important group of tailored graphene derivatives achievable using ultrasound-assisted techniques. Carbon nitrides, in particular, have garnered significant interest due to their remarkable optical and electronic properties [90,91]. Cui et al. [92] compared the thermal acidic/basic peeling of bulk carbon nitride with the one induced by ultrasound-assisted techniques, discussing the current state of the art in the field. The authors reported similar properties and quality achievable with

both approaches but considerably reduced time when using ultrasound-assisted techniques. Huang et al. [93] demonstrated that ultrasound-assisted techniques can not only reduce the size of carbon nitride but also tailor the flakes produced with hydroxyl groups. The resulting materials resembled the structure of GO but exhibited a significant increase in photocatalytic activity due to the tuning of the band gap. As reported by Huang et al. [93], the controlled introduction of defects through ultrasound-assisted techniques allows for precise tuning of the electron conductivity of carbon nitride sheets by simply regulating the stress inside the flakes.

4. UATs for the Production of 2D Materials: Inorganic Materials

4.1. Metal Nitride Species

Metal nitrides (MNs) are inorganic compounds with the empirical formula MxNy, which are produced through the direct reduction of metal oxide or halide at high temperatures using ammonia or nitrogen/hydrogen mixtures [94–96]. Alternatively, nanostructured MNs can be produced using sol-gel techniques, as discussed by Giordano et al. [97]. MNs exhibit several interesting properties, ranging from high hardness to high corrosion resistance and high electrical conductivity [98]. The electronic properties of MNs make them efficient photocatalysts for water pollutant removal [99], and their surface chemistry makes them suitable for adsorptive [100] and filtration systems [101]. Therefore, the production of 2D MN flakes offers a solid solution to amplifying all these properties.

Boron nitride (BN) is one of the most studied MNs due to its similarity to graphene, earning it the acronym "white graphene". BN shares the same hexagonal structure as graphene and has a similar interlayer spacing in multilayer materials (close to 0.333 nm). However, BN is characterized by a very high polarization of the B–N bond, which is not present in the homopolar C–C bonds of graphene [102].

The first study using ultrasound-assisted techniques (UATs) for the production of BN flakes was reported by Han et al. [103]. The authors performed liquid-phase sonication exfoliation of BN single crystals in the presence of a 1,2-dichloroethane solution with a surfactant agent for 1 h. The resulting BN consisted of a mixture of single-layer flakes and multilayer material. This initial attempt at using UATs for the production of 2D BN was further improved by several works that replaced the organic solvent with a water solution of methanesulfonic acid [104]. Štengl et al. [105] employed a mild organic medium environment using dimethylformamide and a high power of up to 2 kW. This approach could be easily scaled up to gram-scale production without substantial modifications [106]. Marsh et al. [107] systematically investigated the effect of various mixtures of water with alcohols and found an optimal trade-off with a 60 wt.% tert-butanol solution. Lin et al. [108] took a step forward by developing UATs for the exfoliation of BN in an aqueous medium for 8 h, suggesting a mechanism as depicted in Figure 3.

The authors of the study reported that sonication initially led to a reduction in the size and number of layers of BN, followed by the cutting of flakes and the production of single-layered BN, as confirmed by transmission microscopy analysis. Interestingly, Deshmukh et al. [109] used plant extracts as a medium for UATs in 2D BN production, demonstrating their ability to act as surfactants.

The use of UATs is not widely employed for the production of other 2D MNs due to the challenges in producing proper flakes. Among the limited works available in the literature, Vasylyev et al. [110] presented an interesting approach for the production of 2D Ti₂N flakes. The authors prepared an alloy of Ti-6Al-4V and submerged it in liquid nitrogen while applying ultrasound sonication at a frequency of 21 kHz and a power of up to 0.6 kW for a maximum duration of 150 s. The material exhibited the formation of small flakes with a few monolayer regions rich in nitrogen, and the presence of MNs was confirmed through XRD analysis. Ruan et al. [111] synthesized VN anchored on carbon nitrides but were unable to isolate the produced MNs, which were significantly larger than monolayer flakes.



Figure 3. Process of exfoliation and size reduction of BN layers. Reprinted with all permission from Lin et al. [108] (Copyright © 2011 American Chemical Society).

4.2. Metal Dichalcogenide Species

Metal dichalcogenides (MDs) are inorganic compounds that consist of a transitional metal ion bonded to chalcogen atoms, such as sulfur, selenium, or tellurium [112]. MDs exhibit interesting electrical and optical properties [113,114], which make them suitable for various environmental applications [115,116]. The production of 2D MDs is possible due to the weak van der Waals forces that occur between the layers of bulk MDs [117].

Among MDs, MoS_2 has been extensively investigated due to its ease of production and remarkable physicochemical properties [118]. Currently, the production of 2D MoS_2 is primarily achieved through physical and chemical vapor deposition techniques, but ultrasound-assisted exfoliation has emerged as a relevant method in recent years [119].

Qiao et al. [120] treated MoS_2 in N-methylpyrrolidone using ultrasound with power ranging from 0.25 to 0.40 kW for 45 min, evaluating the effect of both stable and inertial cavitation in the NMP solvent. The authors demonstrated that low-power ultrasound

promoted stable cavitation, leading to the destruction of Mo–S bonds and overcoming the interlayer interactions, resulting in the formation of large 2D flakes. Higher ultrasound powers induced the formation of smaller flakes due to inertial cavitation. Zheng et al. [121] developed a useful ultrasound-assisted exfoliation method that produced large, high-quality 2D MoS₂ flakes by exfoliating in the presence of hydrazine, naphthalene, and cations under moderate sonication conditions and at a constant temperature.

Das et al. [122] used ultrasound to exfoliate MoS₂ in isopropanol at 0.1 kW power and a frequency of 80 kHz while maintaining a constant temperature of 50 °C. The authors successfully achieved the exfoliation of powdered MoS₂ without inducing any distortion in the crystal lattice structure. Furthermore, they demonstrated the superior performance of $2D \text{ MoS}_2$ over the bulk material in the photodegradation of dyes. Yang et al. [123] applied ultrasound for several hours using a power of 0.5 kW to prepare a stable suspension of 2D MoS₂ flakes in N-methylpyrrolidone for the fabrication of water purification membranes. Smith et al. [124] reported a green route for exfoliating large amounts of MoS_2 in a sodiumcholate-containing water medium using ultrasound with a power of 0.5 kW for up to 480 s and with varying concentrations of surfactants. While this method achieved an inferior performance compared to traditional intercalation methodologies, it demonstrated the effectiveness of ultrasound-assisted exfoliation. Alternatively, Deng et al. [125] used similar conditions but added bovine serum albumin, which acted as both a surfactant and a tailoring agent. The resulting 2D MoS₂ sheets were utilized as a drug delivery system, showing good biocompatibility. Interestingly, ultrasound can also be used to assemble 2D MoS_2 into quasi-zero dimensional particles known as quantum dots, as reported by Gopalakrishnan et al. [126]. The authors demonstrated that sequential sonication steps in a bath and using a tip sonicator induced the formation of 1–2 nm sized particles with enhanced fluorescence emission. Other sulfide-based MNs of great interest include WS₂ [127,128], ReS_2 [129], and NbS_2 [130], which behave similarly under ultrasound irradiation. WS_2 is another interesting case of application for the use of UATs for the production of nanometric materials that has been studied since the late 1980s [131]. WS_2 can be easily exfoliated in water media, obtaining a few layered species [132] with properties close to those displayed by graphene-related materials [133]. Nevertheless, the exfoliation of WS_2 is complex due to the competitive formation of quantum dots together with 2D structures [134].

Similarly, selenides containing mono- and few-layered MNs can be produced using similar routes, particularly for electrochemical applications [135]. Liu et al. [136] reported ultrasound-mediated exfoliation of MoSe₂ in water, bypassing the Hansen solubility parameters [137] through prolonged high-power sonication. The resulting monolayer material exhibited superior performance in the electrochemical hydrogen evolution reaction. Additionally, ultrasound-assisted exfoliation can be combined with cation intercalation approaches to improve the process, as reported by Liu et al. [138].

4.3. MXenes

MXenes are 2D materials discovered in 2011 [139] with the general molecular formula of $M_{n+1}X_n$, where M is an early transition metal and X is a carbon or nitrogen atom [140] with a layered structure. MXenes are generally produced through the strong acidic etching of a precursor with a formula M_nAX_n , where A is an element from mostly the III or IVA groups (i.e., Al, Si, P, S, Ga, Ge, As, Cd, In, Sn, Tl, or Pb) or from groups 13 and 14. The MXenes family has shown remarkable properties for both environmental remediation and pollutant sensing [141,142]. Nonetheless, UATs could be used to further boost the MXenes' performances.

Feng et al. [143] demonstrated that the use of UATs significantly reduced the etching time required for the production of Ti_3AlC_2 by 84%. The authors observed the formation of large-sized flakes with lateral dimensions up to 1 μ m and thickness below 10 nm. This remarkable outcome was attributed to the enhanced heat and mass transfer resulting from local micro-turbulent mixing induced by cavitation collapse. Therefore, careful adjustment of the power can prevent sudden cracks in the basal structure of MXenes while simul-

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taneously improving the etching efficiency. Ghidiu and co-workers [144] proposed that ultrasound irradiation in a water medium facilitated the penetration of water molecules between MXene layers, promoting the weakening of interlayer interactions and consequently leading to the delamination and swelling of the MXenes' original structures [145]. To prevent oxidation during the UAT treatment of MXenes, Mashtalir et al. [146] successfully produced Nb₂C with large lateral sizes under an inert atmosphere. To achieve an oxygenfree environment, it is strongly recommended to degas the reaction mixture through a pre-treatment pulsing ultrasound. However, a poorly optimized process may introduce defects and lead to the formation of metal halide clusters [147], as well as structural and morphological defects that can reduce the performance of MXenes [148].

Another critical parameter to consider is the use of intercalating agents to further enhance the delamination process of MXenes. Naguib and co-workers [149] treated Ti_3CNF_x with water under ultrasound irradiation in the presence of organic amines, which significantly reduced the exfoliation time and fluoride concentration. Wang et al. [150] employed a similar approach using high steric hindrance aryl diazonium salts in water. The authors reported a large and homogeneous distribution of flakes, along with improved colloidal stability of the flake suspension.

Interestingly, as reported by Li et al. [151], UATs can be used to simultaneously produce and exfoliate MXenes. The authors simply used a mixture of Al and TiO_2 suspended in water in an ice bath for 30 min, obtaining a few layer films with a very low resistivity. However, UATs introduced significant changes in morphology and point defects into the structure of MXenes, altering their properties in an uncontrolled way.

5. Advantages and Disadvantages of UATs: A Comparative Discussion

As reported in Table 1, UATs are not the only route to downsizing the materials to 2D dimensions and still face several unresolved challenges. The main issue related to the deployment of UATs is related to the assembly and scale-up of reactors, particularly those operating in continuous flow conditions [152]. As discussed by Meroni et al. [14], the design of UAT reactors is complex: typically they consist of a microfluidic device coupled with an ultrasonic transducer, which can be a piezoelectric plate-based reactor or a Langevin-type transducer, as it requires identifying the reactor's resonance frequency. The transducer optimization is crucial for achieving an efficient setup and involves intricate fluid dynamics calculations, which inherently have limitations due to the combination of various geometries and operating variables (i.e., frequency, power, cycle, reactor vessel/transducer diameter ratio, number, geometry of transducers, and reactor shape). As reported by Tyurnina et al. [153], the production of few-layered graphene can be achieved by using a 2 cm sonotrode at 20 kHz paired with a 3 cm oscillating membrane at 1174 kHz. Nevertheless, the material produced is still confined to the range of a few mg. Asakura et al. [154] scaled up a simple cylindrical reactor to 500 mL, optimizing the sonochemcial efficiency. Nevertheless, the authors aimed to produce a reactor for synthesis and there is still a lack of application for simple exfoliation. Similarly, Nickel et al. [155] developed a reactor with a working volume of up to 29 L optimized for synthesis but adaptable for exfoliation.

Additionally, challenges persist in terms of corrosion resistance and process monitoring for the practical implementation of UATs [156,157]. Alternative approaches with better scalability, such as hydrothermal methods, although less energy-efficient compared to UATs, or microwave-assisted techniques (MATs) [158], may be considered. MATs are similar to UATs but rely on materials that possess good microwave absorption properties, thereby facilitating heat dissipation [159]. Furthermore, MATs possess the same issues regarding parameter control as the wet approaches.

Another approach to address the production of 2D materials is through chemical vapor deposition (CVD) and physical vapor deposition (PVD) techniques [9,10]. CVD and PVD are powerful methods for producing nearly-2D materials with controlled chemistry and good quality. However, their scalability to an industrial level is complex, as several

case studies have demonstrated. PVD, for example, is limited to producing only a few square centimeters/grams per hour [160], and a separation/purification process is often required to remove the support used for deposition. CVD can be applied for the large-scale production of nanomaterials such as carbon nanotubes [160], but it faces similar limitations to PVD when it comes to 2D material production, including the presence of a support material and the generation of side products due to precursor decomposition [161]. Furthermore, CVD does not show the control achievable with other techniques in the production of multilayered materials.

Technique	Advantages	Disadvantages
UATs	 Fast process. Effective on small and medium scales. Energy efficient. High-quality materials produced. Highly tuneable. 	 Hard to scale. Use of solvents. Recovery process required.
MATs	 Fast process. Effective on small and medium scales. Energy efficient. 	 Hard to scale. Mixed-quality materials produced. Temperature control. Required microwave absorbers in the reaction medium. Use of solvents. Purification process required.
Hydrothermal methods	Simple to scale up reactors.Tuneable processes.	 Mixed-quality materials produced. Use of solvents. Purification process required. High pressure and high temperature required.
CVD	 Solventless. Good-quality materials produced. Simple to scale up reactors. Tuneable processes. 	 Support removal. Difficult purification procedures of materials produced. High temperature. High purity of chemicals used during the process.
PVD	 Solventless. High control of 2D material growth. High-quality materials produced. 	Support removal.High temperature.Difficult scalability.

 Table 1. Comparison between techniques used for the production of 2D materials.

6. Conclusions

The production of 2D materials represents one of the more advanced frontiers of material science research. UATs are playing a key role in the advancement of materials production techniques due to their superb result in the downsizing of plenty of different layered species from carbon to inorganic ones. We firmly believe that UATs are a precious toolbox for accessing the nanoworld and for achieving complete control of the tailoring process of 2D materials. Disgracefully, the high energy efficiency and fast process times are counterbalanced by a complex realization of large-scale reactors. This has represented the main issue of UATs for decades, slowing down the spread of these approaches. Nevertheless, the significant technological advancement in the geometry and efficiency of transducers are opening new paths for the development of UATs. Furthermore, the bottleneck represented by the limited and high-cost uses of 2D materials prevents the achievement of the full potential of UATs for the exfoliation and synthesis of 2D materials.

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