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PREPARATION OF THIN FILMS FROM MULTI-LAYER 2D NANOMATERIALS LIQUID

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ABSTRACT:

Thin films comprised of laminated 2D nanomaterials have garnered considerable attention and hold significant promise across a multitude of applications, driven by their exceptional and distinct attributes. This study presents an innovative approach for fabricating thin films on glass substrates through the utilization of 2D multilayer liquid nanomaterials via a sputtering technique. These nanomaterials, encompassing graphene, MoS2, and h-BN, were synthesized employing a common household blender, followed by dispersion in a range of solvents, including acetone, ethylene glycol, and deionized water. In-depth characterization of the resultant thin films was undertaken, employing a comprehensive suite of microscopic and spectroscopic analytical methodologies. The outcomes of these characterizations underscore the profound influence of both the selected bulk material and the choice of solvent on the structural and morphological properties of the thin films. Impressively, the prepared thin films exhibit superlative adhesion and uniformity, manifesting smooth surfaces and a remarkable degree of crystallinity. Through the judicious selection of diverse bulk materials and solvents, this study effectively demonstrates the unparalleled versatility and adaptability of the sputtering method in producing thin films with a wide spectrum of tailored properties. This research not only advances our understanding of thin film formation but also underscores the immense potential of 2D nanomaterial-based films in diverse technological applications.

KEYWORDS: Liquid Exfoliation, 2D Nanomaterials, Thin Films, graphene, MoS2, h-BN.

1. INSTRUCTION

Two-dimensional (2D) nanomaterials exhibit distinctive attributes when compared to alternative nanomaterial forms. Despite their confinement within the 2D nanomaterial structure, charge carriers possess the liberty to traverse along the plane, rendering these substances appealing for fundamental investigations in condensed matter physics and practical implementations in electronic devices (Novoselov et al. 2012). The thickness of these 2D nanomaterials can be precisely manipulated to finely tune their electronic characteristics, a capability absent in other nanomaterials. Furthermore, their minimal thickness provides them with mechanical pliability and frequently results in optical transparency, both of which offer advantages in the development of transparent and flexible optoelectronic devices (A. H. Khan et al. 2017).

Back in 2004, Andre Geim and Konstantin Novoselov achieved a significant breakthrough by successfully isolating and delving into the first 2D material, which they named Graphene (Goni, Chemelli, & Uhlig 2021). Graphene comprises carbon atoms in a sp2 hybridized arrangement, forming a honeycomb structure. This material serves as the foundational element for various carbon-based structures such as carbon dots (0D), carbon nanotubes (1D), and graphite (3D) (Tiwari et al. 2019). Despite its appealing characteristics and promising applications, Graphene's limited band gap has imposed constraints on its utilization in electronic devices. Nevertheless, its exceptionally thin and distinct 2D attributes have propelled it to become the most extensively explored nanomaterial.

In recent times, there has been a growing fascination with the exploration of alternative 2D materials, encompassing hexagonal boron nitride (h-BN) and transition metal dichalcogenides (TMDs), including MoS_2, TiS_2, TaS_2, WS_2, and more (Chhowalla et al. 2013; Huang, Zeng, & Zhang 2013; M. H. Khan et al. 2017; Nicolosi et al. 2013). Among these, molybdenum disulfide (MoS_2) stands out as a 2D material sharing some similarities with graphene but distinguished by its unique mechanical, electrical, thermal, and optical properties. These distinctive features open doors to applications in catalysis, electronics, photonics, energy storage, and sensing (An et al. 2017; Benson et al. 2015; De-Mello et al. 2017; Devices et al. 2014; Li et al. 2016; Ma et al. 2016; Shokri & Salami 2016; Q. H. Wang et al. 2012; Zhu et al. 2017).

2D nanomaterials offer a set of distinct advantages over other nanomaterials. Their ultra-thin, two-dimensional structure provides a remarkably high surface area-to-volume ratio, facilitating enhanced reactivity and catalytic performance. These materials often boast tunable electronic and optical properties, allowing for precise customization to meet specific application requirements. Their exceptional mechanical strength and flexibility make them suitable for applications demanding robust materials, such as flexible electronics and reinforced composites. Additionally, some 2D nanomaterials exhibit outstanding electrical conductivity, making them invaluable in high-performance electronic components. These unique properties, coupled with their biocompatibility, barrier properties, and environmental benefits, position 2D nanomaterials as versatile and promising candidates for a wide range of technological innovations.

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Hexagonal boron nitride (h-BN), on the other hand, is characterized by covalent bond planes held together by weaker Van der Waals forces. This inherent fragility makes it susceptible to peeling into single-layer or polyatomic nanosheets (BNNSs), with lateral dimensions typically spanning hundreds of nanometers. The 2D h-BN nanosheets have been anticipated to possess a substantial band gap, triggering significant research interest and inspiring the quest for ultra-thin semiconductor materials at the atomic scale (Dlouhy, 2021).

Over the course of time, a multitude of techniques have been developed to fabricate 2D nanomaterials, broadly categorized into two approaches: top-down and bottom-up methods (Coroş et al. 2019). Top-down methods encompass the separation of stacked graphite layers to yield graphene sheets, achieved through various processes such as mechanical exfoliation, liquid phase exfoliation (LPE), and arc discharge (Balasubramanyan et al. 2017; Poorali & Bagheri-Mohagheghi 2017; Smovzh et al. 2017; Y.-Z. Wang et al. 2018; Yao et al. 2012; N. Zhang et al. 2017). In contrast, bottom-up methodologies involve the synthesis of graphene from carbon-rich sources, employing techniques such as chemical vapor deposition, epitaxial growth, and pyrolysis (Lin et al. 2019; M'Barki, Bocquet, & Stevenson 2017; Teeter et al. 2017; H. Zhang et al. 2019).

Recently, a novel approach called liquid exfoliation has emerged as an effective means of generating graphene nanosheets. This process involves subjecting graphite to high shear rates, achieved through either a high-shear rotor-stator mixer or a basic graphite mixer (Jafarpour et al. 2021; Paton et al. n.d.; Varrla et al. 2014; Varrla, Backes, Paton, Harvey, Gholamvand, Mccauley, et al. 2015). Notably, this technique exhibits scalability advantages, especially for handling large volumes. However, its adaptation for inorganic layered materials remains in the early stages of development (Paton et al. n.d.; Varrla, Backes, Paton, Harvey, Gholamvand, Mccauley, et al. 2015). Ongoing endeavors are focused on fine-tuning process parameters and expanding the use of blade mixers to scale up the liquid phase exfoliation of these materials (Yi & Shen, 2014).

In recent times, 2D nanomaterials have garnered considerable attention, driven by their distinctive attributes and wide-ranging utility These materials, which encompass MXenes, graphene, transition metal dichalcogenides, and black phosphorus, exhibit a host of compelling features, including robust electrical conductivity, mechanical resilience, and the ability to fine-tune their band gaps. These characteristics render them exceptionally adaptable for a multitude of technological applications. The applications of 2D nanomaterials extend across a spectrum of domains, encompassing agriculture, electronics, energy harvesting, and biomedicine. In the realm of agriculture, nanomaterials find purpose in safeguarding crops, manipulating plant genetics, and enhancing food production and safety. Within electronics, 2D nanomaterials play an integral role in shaping nanoscale semiconducting devices, optoelectronics, and micro-electro-mechanical systems. Meanwhile, energy harvesting technologies, exemplified by triboelectric nanogenerators and piezoelectric nanogenerators, harness the exceptional qualities of 2D materials for enhanced performance. Furthermore, in the field of biomedicine, MXene-based materials have exhibited notable potential. They have demonstrated efficacy in diverse applications, spanning from drug delivery systems to the immobilization of noble metal nanoparticles, opening new avenues for innovation. These manifold and varied applications underscore the profound significance of 2D nanomaterials across a multitude of disciplines, emphasizing their role as a fertile ground for ongoing research and development endeavors (Khandelwal et al. 2023; Sachan et al. 2023; Siddappa A. Patil, Kostiantyn O. Marichev, Shivaputra A. Patil n.d.).

In this particular research study, a variety of 2D nanomaterials were prepared using a straightforward and cost-effective approach involving shear exfoliation methods, specifically employing a Home Blender to ensure high quality. As a proof of concept, the resulting liquid materials were subsequently utilized to create thin films through a spray coating process, with a focus on exploring their potential applications in optoelectronics.

2. Materials and Methods

All chemicals and solvents were utilized as received without undergoing any further purification. Graphite powder, MoS_2, and h-BN were purchased from Aldrich, while acetone (CH_3COCH_3) and ethylene glycol ($C_2H_6O_2$) were acquired from Sober Life Co. Additionally, various other materials such as olive oil, stone, Halab's soap, and dishwashing liquid were procured from commercial sources.

To disperse and exfoliate the materials in bulk form, a common kitchen blender plays a remarkable role in preparing high-quality few-layer graphene (FLG) and multi-layer 2D nanomaterials. This unassuming appliance induces intricate fluid dynamics, including shear forces, turbulence, and particle collisions, facilitating the exfoliation of graphite into exceptional FLG flakes. This innovative method, driven by the blender's shear-assisted exfoliation, has emerged as a leading technique for graphene synthesis, particularly in photocatalytic applications. A kitchen blender (Model GSB-426) was employed, utilizing different solutions including deionized water, ethylene glycol, and acetone. Firstly, the required amount of surfactant was dissolved in DI water through stirring. Subsequently, the loose materials were added to a conical mixing bowl, and the aqueous solution was poured on top. The mixer operated at a fixed speed for a specific duration, but due to heat concerns, a 5-minute blending cycle was followed by a 1-minute off cycle, with the flask placed in an ice bath during the shutdown. After blending, the supernatant was carefully decanted and subjected to ultrasonic bath treatment for an hour. The mixture was then sonicated for an additional hour. Once an optimized solution was determined, a thin film of multiple 2D nanomaterials on a glass substrate was prepared using the sputtering coating method. The substrate was then left to dry at room temperature for several hours, allowing the solvent to evaporate and leaving behind a thin film of multi-2D nanomaterials on the glass surface. Table 1:1 displays a juxtaposition of diverse investigations on exfoliation using solvent-based techniques. The resultant fluid mixture yielded by liquid-phase exfoliation (LPE) finds utility across a spectrum of domains, encompassing coatings, thin-film sectors, and has exhibited impressive outcomes in a myriad of uses like battery cathodes, fiber lasers, and fortified composites (Chaudhary, Bansal, & Khanuja, 2021).

In the realm of thin film preparation on glass substrates, several methods have been developed to achieve uniform and

precise deposition. These techniques include PVD, CVD, sol-gel, spin coating, and spray coating. Among these methods, spray coating emerges as the most advantageous for several reasons. Spray coating involves atomizing a solution or suspension into fine droplets, which are then sprayed onto the glass substrate. This technique provides superior control over film thickness, uniformity, and coverage, making it ideal for large-scale production with consistent results. Moreover, spray coating allows for the deposition of various materials, making it versatile for different applications, from protective coatings to optical films. Its ease of implementation, cost-effectiveness, and adaptability to irregularly shaped substrates further solidify spray coating as the best method for preparing thin films on glass substrates.

In the preparation of multi-layer 2D nanomaterials using a blender, the choice of solvents like deionized water, acetone, and ethylene glycol plays a pivotal role. Among these options, pure acetone emerges as the optimal solvent for dispersing and exfoliating graphite, MoS_2, and h-BN nanomaterials. Acetone's exceptional ability to rapidly evaporate and effectively disperse these nanomaterials contributes to the production of high-quality solutions, facilitating subsequent thin film fabrication.

The obtained thin films underwent characterization using various techniques. The morphology of the samples was analyzed using a scanning electron microscope (SEM) (Quanta 450). The atomic composition of the elements was confirmed by energy-dispersive X-ray (EDX) spectroscopy, performed at SEM. Furthermore, the quantum properties of the thin films were evaluated at room temperature through X-ray diffraction (XRD) measurements using a PAN X' Pert PRO analyzer (Cu Kα = 1.5406 Å), which allowed the examination of possible phase and crystallinity, along with radiograph recording spectrophotometry. The absorption spectra of the 2D nanoscale multi-material thin films prepared on a glass substrate were studied using visible ultraviolet light (Shimadzu UV-160A).

Table 1. Evaluating the Exfoliation of 2D Materials through Solvent-Based Liquid Phase Techniques.

S. no.	Material	Solvent	Exfoliation time (min)	Ref.
1	WS_2	Ammonia	180	(Adilbekova et al. 2020)
2	$MoSe_2$	Anhydrous ethanol	600	(Chen et al. 2019)
3	Graphitic flake	NMP	30	(Hernandez et al. 2008)
4	Boron nitride (BN)	Urea with glycerol (2:1)	1440	(Zheng et al. 2019)
5	Black phosphorous (BP)	CHP	300	(Hanlon et al. 2015)
6	MoS_2	NMP, DMSO, and Ethanol	10, 20, 30, 40, 50 and 60	(Chaudhary, Bansal, and Khanuja 2021)
7	Graphite flake, MoS_2 and h-BN	*Deionized water*, Acetone and *Ethylene glycol*	50 and 60	Presented work

Thin films can be crafted using diverse methodologies. One approach entails utilizing specialized equipment designed for thin film fabrication, where a volatile solvent, blended with a film-forming substance, is employed. This equipment undergoes rotational motion to flatten and evaporate the solvent, while a fixture secures the substrate targeted for coating (Wang Chunrui, Chen Xing, Junfeng Shao n.d.). In a different context, the creation of a thin-film battery involves a multi-step sequence. This intricate process encompasses dry etching a silicon wafer to shape microcell patterns, introducing doping to these patterns, transferring them onto a polyamide layer, applying a conductive layer via ink-jet printing, and finally, laminating to construct a flexible back plate (Liu Zhuo, n.d.). Additionally, in the domain of thin-film solar cell production, a method is employed wherein an initial covering layer is applied to a resin film layer through a coating process, followed by the deposition of a secondary covering layer, composed of an inorganic material. This technique enhances the thin film's water resistance and, consequently, the overall durability of the thin-film solar cell (Xu Qiang, Bai Anqi n.d., n.d.). However, among these methods, spray coating emerges as the most advantageous choice for a multitude of reasons. This technique revolves around the atomization of a solution or suspension into fine droplets, which are subsequently sprayed onto the glass substrate. The notable benefits of spray coating encompass superior control over film thickness, uniformity, and coverage, rendering it an optimal choice for large-scale production characterized by consistency. Furthermore, spray coating demonstrates adaptability in accommodating various materials, rendering it versatile for an array of applications, ranging from protective coatings to optical films. Its simplicity of implementation, cost-efficiency, and suitability for irregularly shaped substrates further establish spray coating as the preeminent method for thin film preparation on glass substrates.

3. RESULTS AND DISCUSSION

Direct observation studies have provided valuable insights into the optimal preparation liquid for obtaining high-quality multi-2D nanomaterials from graphite, MoS_2, and h-BN as bulk materials powders using a blender. Among the tested solutions, acetone has been identified as the most effective choice for preparing multi-2D nanosheets. Acetone enables efficient dispersion and exfoliation of the bulk powders, successfully forming multi-2D nanomaterial structures. In contrast, ethylene glycol and deionized water have demonstrated limited effectiveness in achieving high-quality multi-2D nanosheets. While ethylene glycol promotes some degree of exfoliation, it falls short of producing the same level of quality and yield as Acetone. Deionized water, on the other hand, exhibits limited exfoliation and dispersion capabilities for bulk powders, making it less suitable for the production of multi-2D nanomaterial. Thus, direct observation indicates that acetone emerges as the optimal preparation liquid for obtaining high-quality multi-2D nanomaterial from bulk powders using a blender (Fig. 1).

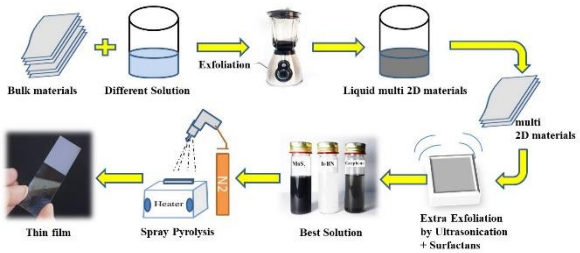


Figure 1: Schematic representation of thin film preparation

Figure placement and numbering

The preparation of thin films from liquid multi-2D nanomaterials, such as graphene, MoS_2, and h-BN, can be achieved by spray coating on glass substrates with heaters. In this method, a liquid solution containing the dispersed nanosheets of the desired nanomaterial is loaded into a spray coating gun. The gun emits a fine mist of the solution onto the glass substrate, which is positioned on a heater. The heater ensures a controlled temperature, promoting the evaporation of the solvent and facilitating the formation of a uniform thin film. The solvent evaporates as the mist settles on the heated substrate, allowing the nanosheets to assemble and form a thin film. The combination of the spray coating gun process, the heater, and the glass substrate enables precise and efficient deposition of the nanomaterials, resulting in thin films with tailored properties. This approach holds great promise for many applications, including electronics, optoelectronics, and power, where uniform and controlled thin films are critical to device performance and functionality.

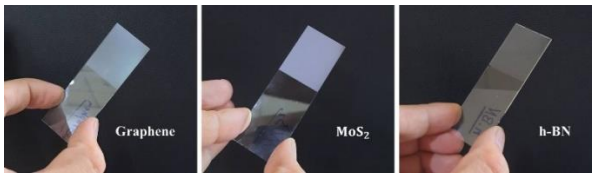


Figure 2: Thin film prepared by spray coating

To study the morphology of the prepared thin films, SEM is used to test the prepared thin films. Figure 3 shows an optical micrograph of the deposited thin film showing the shape and dimensions of the graphene-MoS_2-h-BN sheet-like structure. Graphene nanoflakes acquired are relatively thin and have measurements of a few hundred nanometers. Moreover, the nanoflakes' small thickness is confirmed by the visibility of the substrate's background through them (Fig.3a). SEM images are obtained at various magnifications, ranging from low to high resolution. At low magnifications, the SEM provides an overview of the sample surface, revealing the overall morphology of the graphene's few-layer thin film. The images show the presence of wrinkles, folds, or stacking defects in the film.

MoS_2 film (Fig.3b) appeared flat and homogeneous, indicating successful exfoliation and deposition. The arrangement and distribution of individual MoS_2 flakes were observable, providing insights into the film's uniformity and coverage. In the case of h-BN few layers of thin films, SEM analysis reveals the film's overall surface topology with h-BN hexagonal structure, corresponding to the individual layers of h-BN (Fig.3c).

In summary, SEM analysis reveals that all the prepared thin films have no irregularities in high-quality deposition, and the surfaces are uniform, with minimal roughness. However, a few defects like cracks, pinholes, and particle contamination also be observed. Furthermore, correlating the imaging contrast and intensity variations with calibrated standards made it possible to determine the approximate number of MoS_2 layers present in the film. The results indicated a few-layer thickness, consistent with the exfoliation method used. In addition, EDS mapping of chemical elements present in the film, confirms the presence of C, M, O, S, B, and N atoms (Fig.3 d, e, and f) respectively.

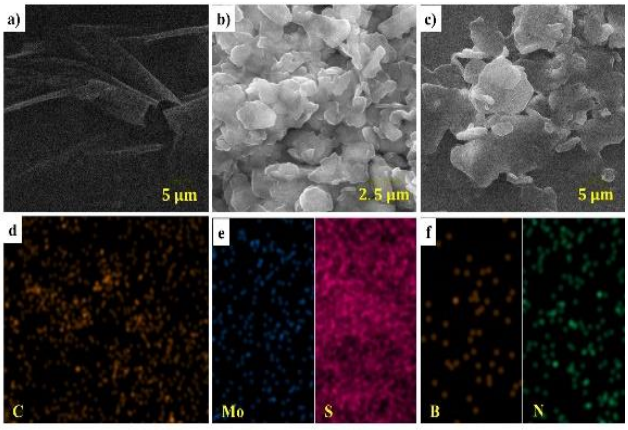


Figure 3: SEM images for thin films as pure 2D materials a) graphene, b) MoS2, and c), h-BN

The employed methods, including liquid preparation and thin film production, provide strong resilience and can be further utilized for various purposes. Exfoliated nanomaterials can be utilized to create diverse composite liquid substances. The prepared composite thin film is depicted in Figure 4. As displayed in Figure 4a, h-BN nanosheets are layered on top of MoS_2 sheets. Likewise, Figure 4b and c exhibit a haphazard arrangement of graphene flakes with stacked MoS_2 and h-BN layers respectively.

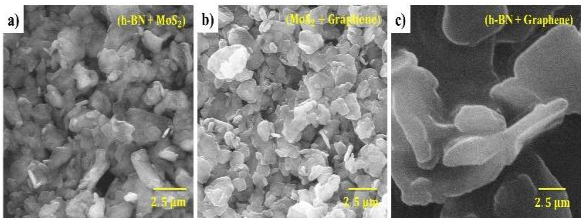
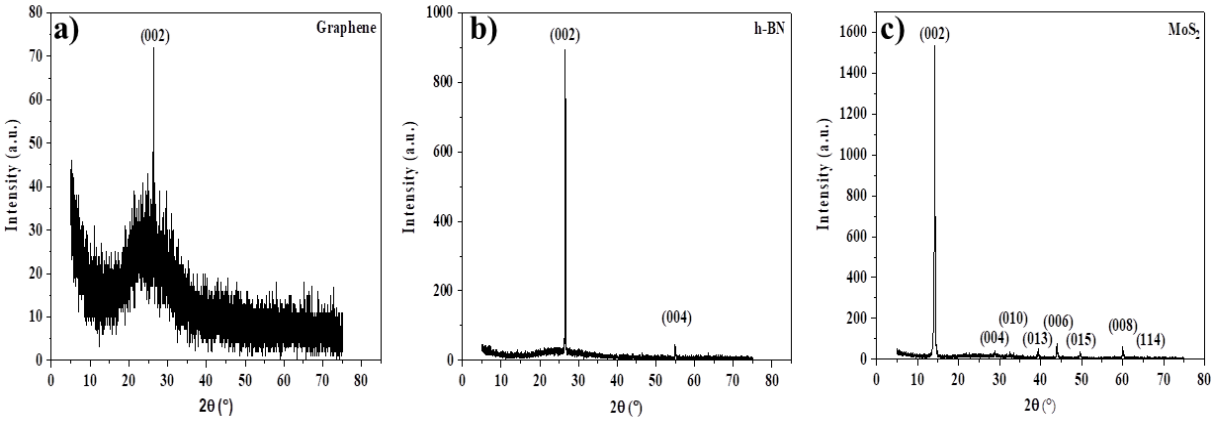


Figure 4:SEM images for thin films composite multi 2D materials

X-ray diffraction (XRD) analysis of graphene, MoS_2, and h-BN thin films allows us to determine their crystal structures. Graphene exhibits a sharp peak at $2\theta \approx 26.5°$, indicating a highly crystalline hexagonal (002) plane of uniform thickness. The intensity of the (002) peak also estimates the number of graphene layers. MoS_2 thin film exhibits multiple diffraction peaks in its XRD pattern, each corresponding to a specific crystallographic direction, providing information about film thickness, crystallinity, and orientation. The intensity and relative positions of these peaks indicate a highly crystalline and well-aligned MoS_2 thin film, impacting its electronic and optical properties. Similarly, h-BN thin films show diffraction peaks, including (002) and (004), revealing information about thickness, crystallinity, and orientation. The (002) peak corresponds to interlayer spacing, while the (004) peak corresponds to the in-plane crystal plane. For each thin film, distinguishing the signal for the (002) plane from the bulk powder confirms the fabrication of ultrathin films with few layers. Multiple peaks suggest a more complex crystal structure, affecting electronic and optical properties.

Figure 5: XRD pattern of the deposited thin film. Diffraction peaks are indexed from the graphene phases (reference code. 75-1621),



MoS_2 (reference code. 024-0513, and h-BN (reference code. 01-071-2649)

Based on the atomic percentages from EDX analysis (Fig. 6), the prepared graphene thin film contains a significant amount of carbon, with an atomic percentage of 31.55%. Also, it appears that the h-BN thin film contains a relatively high atomic percentage of boron (26.5%) compared to nitrogen (5.29%). For MoS_2 thin film, it contains a relatively low atomic percentage of molybdenum (8.38%) compared to sulfur (34%) and oxygen (37.33%). The presence of oxygen in each graphene, MoS_2, and h-BN thin film could be due to a few different factors. One possibility is that the sample was exposed to air during preparation or analysis, resulting in the oxidation of the surface.

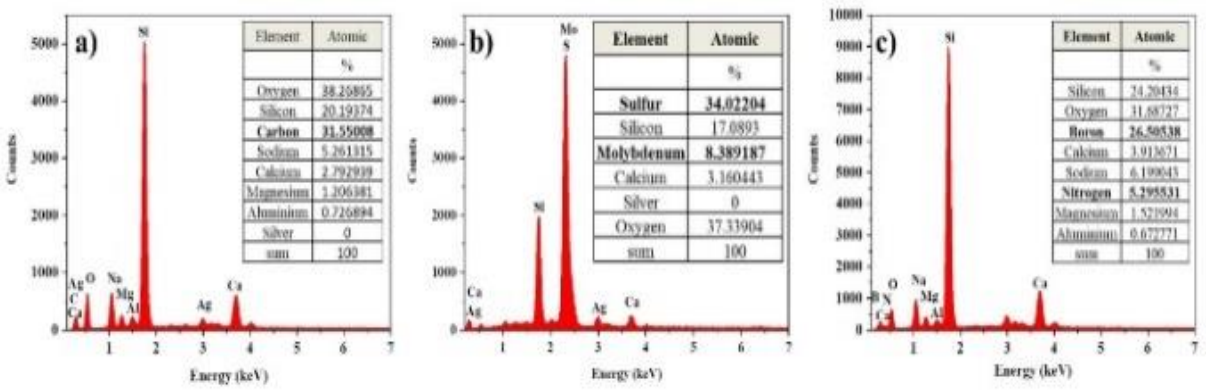


Figure 6: EDX analysis of the a) Graphene b) MoS_2 and c) h-BN thin films along with the elemental composition. The inset of each figure is the atomic percentages

To investigate the optical properties of thin films obtained from stable dispersions of graphene, MoS_2, and h-BN, produced through shear exfoliation using a mixer for thin film preparation by spray coating, a comprehensive range of transmittance measurements was conducted across the spectrum of 200–1200 nm (Fig. 7a). The bandgap energy (E_g) of the prepared thin films was determined by analyzing their transmittance curves. For this analysis, we plotted the photon energy (hv) against the square of the product of the absorption coefficients of the photon energy $(\alpha hv)^2$. The E_g values were obtained by extrapolating the linear part of the $(\alpha hv)^{1/2}$-hv curve to the corresponding horizontal B-band photon energy axis, applying Tauck's law (Varrla, Backes, Paton, Harvey, Gholamvand, McCauley, et al., 2015; Yi & Shen, 2014) (Fig. 7b).

The evaluation of energy gap values in multilayer graphene thin films revealed a measure of 2.5 electron volts (eV), signifying the transition from the valence band to the conduction band. While single-layer exfoliated graphene manifests a distinct bandgap (Yi & Shen, 2014), bilayer graphene, owing to the asymmetry in on-site energies between its layers induced by an external gate, has the potential to introduce gaps between the otherwise degenerate graphene layers, theoretically establishing both conduction band and valence band. A comparable energy gap is expected to emerge in multiple graphene layers, with the gap's magnitude contingent upon the layer count (Varrla, Backes, Paton, Harvey, Gholamvand, McCauley, et al., 2015). Additionally, the indirect energy gap values (E_g) for MoS_2 and h-BN thin films were computed at approximately 2.2 eV and 3.64 eV, respectively, as illustrated in Figure 7b. Notably, the high E_g value for h-BN underscores its insulating characteristics.

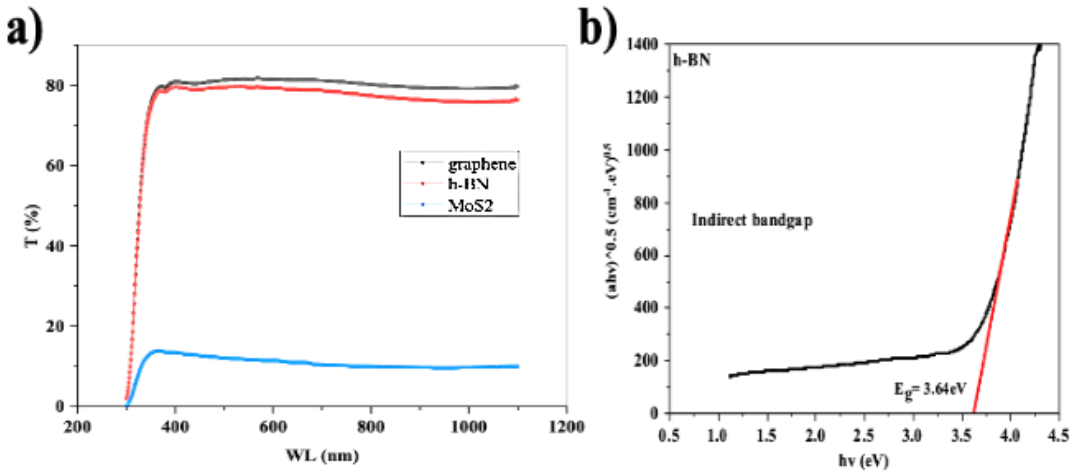


Figure 7: Transmittance spectra for different multi-2D material thin films

4. CONCLUSION

To summarize, multi-layer liquid 2D nanomaterials can be easily prepared from bulk graphite, MoS_2, and h-BN using a home blender in various solvents such as di-water, acetone, and ethylene glycol. This method has shown promising results in thin film production through spray coating. The use of a home blender is a cost-effective and simple approach that can be implemented both in laboratories and at home. Choosing the right solvent is crucial for achieving high-quality thin films, with acetone being the most effective option. Spray coating allows for convenient and scalable production of large-area multi-2D nanomaterial films.

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