HORIZON EUROPE PROGRAMME
HORIZON-CL4-2023-DIGITAL-EMERGING-01-33

GA No. 101135196

Developing New 2D Materials and Heterostructures for Printed Digital Devices

2D-PRINTABLE - Deliverable report

D3.1. – Ink production/printing

Funded by
the European Union

Deliverable No.	D3.1	
Related WP	WP 3	
Deliverable Title	Ink production/printing	
Deliverable Date	2024-09-30	
Deliverable Type	REPORT	
Dissemination level	Public (PU)	
Author(s)	Joka Buha (BeD) Cian Gabbett (TCD) Jonathan Coleman (TCD) Sebastiano Bellani (BeD)	2024-09-09
Checked by	Jonathan Coleman (TCD)	
Reviewed by	Sebastiano Bellani (BeD) Ali Shaygan Nia (TUD) Paolo Samori (UNISTRA)	
Approved by	Jonathan Coleman (TCD) - Project Coordinator	
Status	Draft v1	2024-09-09

Document History

Version	Date	Editing done by	Remarks
V1.0	2024-09-03	Joka Buha	TCD contribution, dated 24-08-2024, was included here
V1.1	2024-09-04	Sebastiano Bellani	
V2.0			
V3.0			
FINAL			

Project Scientific Abstract

The 2D-PRINTABLE project aims to integrate sustainable large-scale liquid exfoliation techniques with theoretical modelling to efficiently produce a wide range of new 2D materials (2DMs), including conducting, semiconducting, and insulating nanosheets. The focus includes developing the printing and liquid phase deposition methods required to fabricate networks and multicomponent heterostructures, featuring layer-by-layer assembly of nanometer-thick 2DMs into ordered multilayers. The goal is to optimize these printed networks and heterostructures for digital systems, unlocking new properties and functionalities. The project also seeks to demonstrate various printed digital devices, including proof-of-principle, first-time demonstration of all-printed, all-nanosheet, heterostack light-emitting diodes (LEDs). In conclusion, 2D-PRINTABLE will prove 2D materials to be an

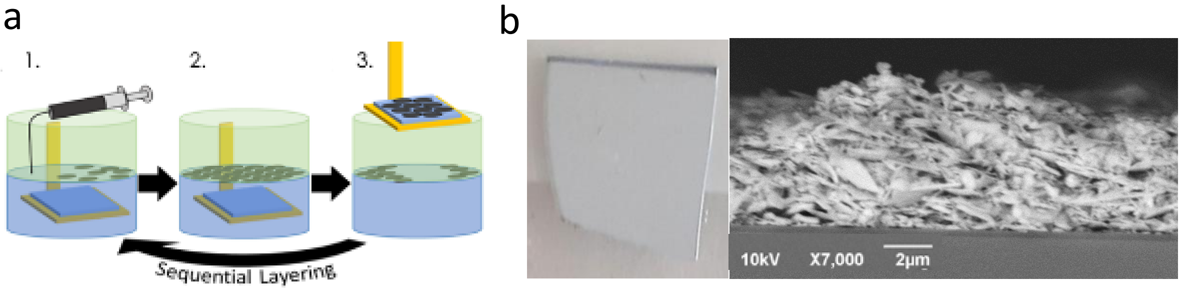
indispensable material class in the field of printed electronics, capable of producing far-beyond-state-of-the-art devices that can act as a platform for the next generation of printed digital applications.

Public summary

This deliverable (D3.1) report outlines significant progress in developing cutting-edge 2D materials for next-generation printed electronics. Our work focuses on improving the quality and performance of these materials by refining how we prepare and apply them. Key achievements reported in this document include:

1. **Production of novel 2D materials through liquid-phase exfoliation techniques**: we explored various 2D materials, including conductive electrochemically exfoliated graphene, semiconducting transition metal chalcogenides, and dielectric oxyhalides, refining them towards newly tailored properties.
2. **Optimization of novel 2D material ink formulations**: by adjusting ink properties such as viscosity and solvent composition, we ensured that these inks are suitable for high-quality printing and coating.
3. **Validation of printing techniques for 2D material inks**: several deposition methods, including Langmuir-Schaeffer (LS) liquid-liquid interfacial deposition and spray coating, were screened and optimized. These methods are essential for creating uniform and high-performance films from our 2D material inks, as needed for solution-processed digital electronics

This progress is a crucial step toward creating more efficient, flexible, and cost-effective digital electronic devices. By enhancing the performance of printed electronics, these advancements could lead to new applications in various fields, including wearable technology, smart packaging, and next-generation sensors.



On the left (a), is a schematic of the LS liquid-liquid interfacial deposition process used for the deposition of monolayers of EE produced materials. An example of novel 2D material printed using a spray printing technique is shown in (b). The ink prepared from LPE produced PrOBr forms a compact film on the surface of Si wafer composed of platelets layered on top of each other.

Contents

List of Figures

Abbreviations & Definitions

Abbreviation	Explanation
PVP	Polyvinylpyrrolidone
DMF	Dimethylformamide

EE	Electrochemical exfoliation
LPE	Liquid phase exfoliation
IPA	Isopropanol
DI	Deionised
TMDs	Transition metal dichalcogenides
AgNS	Silver nanosheets
LS	Langmuir-Schaeffer
TMMs	Transition metal monochalcogenides
h-BN	Hexagonal boron nitride
LCC	Liquid cascade centrifugation
NMP	N-Methyl-2-pyrrolidone

1 Introduction

The development of novel two-dimensional (2D) materials and their integration into scalable, high-performance electronic devices represents a significant milestone in the field of printed electronics. This deliverable focuses on the progress made in optimizing and characterizing novel 2D material inks and their deposition techniques, which are crucial for achieving the project's objectives.

The research presented herein explores various types of 2D materials, including conducting, semiconducting, and dielectric nanosheets, produced through different exfoliation methods. Specifically, the deliverable examines the use of electrochemical exfoliation (EE) and ultrasonication-assisted exfoliation to produce graphene and other 2D materials (chalcogenides and oxyhalides), as well as the use non-layered colloidal metallic nanosheets. These materials were selected based on their potential for enhancing the performance of printed electronic devices by offering superior electrical, optical, and mechanical properties compared to conventional materials.

To achieve high-quality, functional coatings and films, it was essential to tailor the stabilization and deposition strategies for these diverse 2D materials. The deliverable details the optimization of ink stability and rheological properties, which are critical for the successful application of high-throughput printing and deposition techniques. Techniques such as Langmuir-Schaeffer (LS) liquid-liquid interfacial deposition, spray coating, spin coating, and aerosol jet printing were employed to process these inks.

The deliverable provides a comprehensive analysis of the inks' preparation, stabilization, and deposition processes, and presents data on the performance and uniformity of the deposited films. It also highlights the improvements made in film thickness homogeneity and the impact of various processing parameters on the final material properties.

Overall, the results reported in this deliverable contribute to the advancement of 2D material technology and its application in printed electronics.

2 Methods

2.1 Background

In this deliverable, different types of two-dimensional (2D) inks were studied: nanosheets produced via electrochemical exfoliation (EE), nanosheets produced via ultrasonication-assisted exfoliation, non-layered colloidal metallic nanosheets. Due to the distinct physical characteristics of these 2D materials, tailored stabilization and printing/deposition strategies were required. The 2D material inks characterised here comprise conducting (graphene, silver nanosheets -AgNSs-), semiconducting (metal chalcogenide nanosheets, and dielectric oxyhalides. The ink stability and rheology were optimized for high-throughput printing/deposition techniques, such as spray-coating, aerosol jet printing, spin coating, and Langmuir-Schaeffer (LS) liquid-liquid interfacial deposition. Based on the characteristics of the nanosheets, focus was directed on some of these techniques, in particular LS liquid-liquid interfacial deposition and spray coating.

2.2 Procedures

Nanosheet inks were prepared through established EE protocols. After exfoliation, the resulting materials were size-selected using liquid cascade centrifugation (LCC) to ensure uniform flake size distribution[2]. The initial solvent matrix for 2D TMD and TMM nanosheets consisted of dimethylformamide (DMF) and polyvinylpyrrolidone (PVP), which was selected for its stabilizing properties. Similarly, EE graphene inks were produced in DMF, forming stable dispersions. Both conducting and semiconducting inks were transferred to isopropanol (IPA) via centrifugation to optimize them for deposition. Additionally, AgNS inks were prepared by diluting a commercially sourced aqueous stock dispersion with deionized (DI) water. The tailored 2D inks were deposited using established protocols for LS deposition[3], spray-coating[4], spin-coating and aerosol jet printing[5].

Lastly, bulk oxyhalides, as well as an additional low-bandgap layered semiconducting chalcogenides (As_2S_3) in powder or small crystal form were dispersed in solvents such as IPA or a mixture of IPA and DI water. Ultrasonication for 8 h was used to exfoliate the materials in an environmentally friendly solvent system, avoiding toxic, high-boiling-point solvents like N-Methyl-2-pyrrolidone (NMP). After exfoliation, the solvent was removed by centrifugation, and the precipitate containing both exfoliated and unexfoliated material was redispersed in a smaller volume of isopropanol (5–10 mL) via further sonication. This step adjusted the ink concentration for spray coating while maintaining the necessary low viscosity (<100 mPa·s). After sedimentation for 2 h, the upper part of the supernatant was collected. No binders or rheological additives were used, though their inclusion will be considered in future studies for other deposition techniques. As-produced oxyhalide inks were deposited using a spray coating gun with a 35 µm nozzle onto various substrates (quartz slide, pyrolytic graphite, or Si wafer) heated to 45°C. Characterisation of the 2D materials and corresponding inks/films was performed using a range of standard techniques such as optical extinction spectroscopy, scanning electron microscopy (SEM), transmission electron microscopy (TEM), X-ray diffraction (XRD), Raman spectroscopy, and atomic force microscopy (AFM) and rheological characterisation.

2.3 Data Analysis

The data was analysed using OriginPro. Analysis of the network thickness homogeneity for LS-deposited EE 2D inks was performed using Python.

3 Results & Discussion

3.1 Results

3.1.1 Improving the ink stability

Producing stable 2D inks of high aspect ratio TMDs and TMMs using electrochemical exfoliation has been the focus of intensive research over the last few years[6]. The use of PVP as a stabilizing polymer is central to this as it helps to maintain the exfoliated state of TMD/TMM layers in the ink, while also preventing reaggregation during size-selection. This is crucial, as it is necessary to employ centrifugation steps at low speeds to remove thick (> 50 nm) nanosheets, and higher speeds to selectively sediment nanosheets with large aspect ratios (> 100). DMF was chosen as the initial solvent for EE TMDs/TMMs due to its high polarity and ability to dissolve PVP effectively, facilitating the dispersion of these materials at the nanoscale. However, for solution processing and deposition, particularly for applications such as printing or spin-coating, a solvent exchange to IPA was performed. IPA was selected due to its lower boiling point and improved wettability on a variety of substrates, which aids in the formation of uniform thin films during the deposition process. Prospectively, these approaches can be used also for the novel 2D materials explored in the project, including dielectric oxyhalide ones and other chalcogenides (As_2S_3), herein screened without any stabilizing polymer to preliminary assess a simple deposition through spray coating, compatible with available material amounts.

3.1.2 Rheological optimisation of EE 2D Material Inks and Printing

The rheological properties of 2D material inks, such as their viscosity, are crucial in determining their suitability for various deposition techniques. In the case of EE TMDs and TMMs, the ink viscosity is influenced by the concentration of PVP, the size distribution and concentration of the nanosheets, and the solvent used. These rheological properties affect the flow and spread of the ink during deposition, but also play a crucial role in defining the final morphology and electrical properties of the deposited films[7, 8]. Therefore, careful control of these parameters is essential to optimise the performance of TMD/TMM-based devices fabricated using these 2D inks. Here, we have utilised an ink viscosity of ~ 1 mPa s to ensure compatibility across the deposition techniques available to us, such as spin-coating, LS deposition, inkjet printing, spray-coating and aerosol jet printing.

In particular, we have worked to optimise the deposition process for EE 2D inks using the LS technique, shown in Fig. 3.1a. Here, the choice of IPA as the solvent matrix results in an interfacial tension gradient at the liquid-liquid interface. This serves to compress the nanosheets into a tightly-packed monolayer with predominantly edge-to-edge nanosheet contacts (**Figure 1**a). Cosolvent ink formulations of DI water/IPA have been developed for LS-deposition of AgNSs. The addition of IPA reduces mixing of the AgNS ink with the DI water subphase and facilitates the formation of tiled AgNS monolayers.

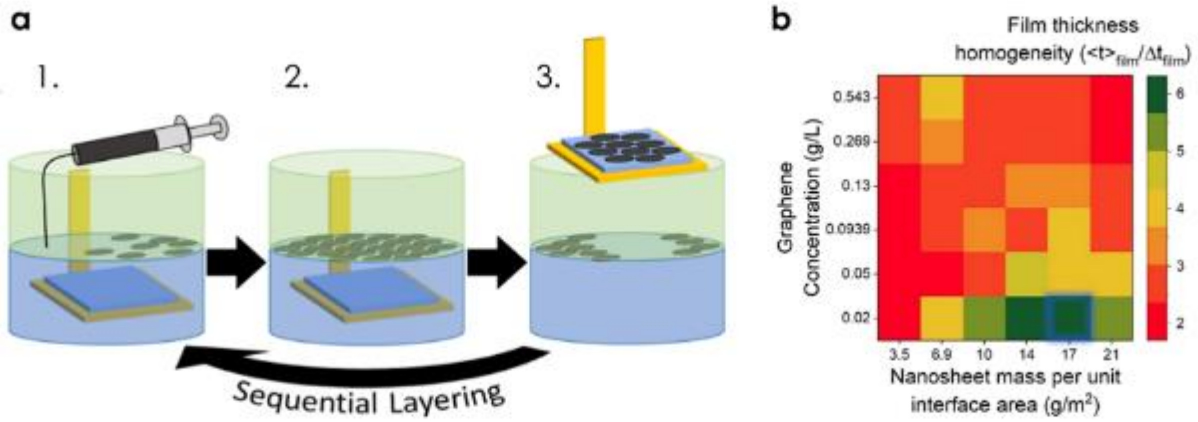


Figure 4. *Optimisation and LS-deposition of EE graphene inks. a) Schematic of the LS liquid-liquid interfacial deposition process. (1) The 2D ink is injected at the water-hexane interface. (2) A complete monolayer of EE graphene nanosheets is formed. (3) The substrate is lifted through the interface to deposit the monolayer. This process can be repeated for thicker films or multiple layers. b) Measurements of the film thickness homogeneity for LS-deposited EE graphene, as a function of ink concentration and the areal nanosheet loading at the liquid-liquid interface. The film thickness homogeneity is described by the ratio of the mean film thickness,*$\langle t \rangle_{film}$*, and the standard deviation in film thickness,* Δt_{film}*, determined from optical scans of each network. Lower ink concentrations lead to improved (larger) values in film thickness homogeneity. Each square in b) represents a LS-deposition over an area of ~ 4 cm*^{2}*.*

Work to characterise the effect of ink concentration on network formation has been performed using LS-deposited EE graphene nanosheets in IPA as a model system. A stock EE graphene ink was diluted to various concentrations in the range of 0.02 – 0.54 g L^{-1}, which were determined using established optical extinction metrics[9]. These inks were then LS-deposited, and the resultant network homogeneity was determined as a function of the ink concentration and nanosheet areal mass loading using optical transmission scans. As shown in Fig. 3.1b, the deposited films exhibit increased homogeneity in film thickness for low-concentration inks (~ 0.02 g L^{-1}). Furthermore, it was determined that the film thickness homogeneity is maximised for injections of ~ 17 g m^{-2} of EE graphene at this ink concentration (Fig. 3.1b). This will inform protocols for optimised deposition of 2D inks, and reduced material wastage, using the LS technique going forward.

3.1.3 Sonication-assisted exfoliation of dielectric oxyhalides and other layered materials

Total of 9 novel materials were exfoliated and used for ink formulation and subsequent spray coating. Some of these inks are shown in **Figure 2**.

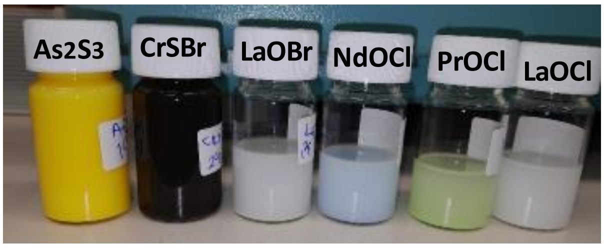


Figure 5*. Examples of novel 2D material inks. The inks are in IPA and produced by sonication-assisted LPE from novel 2D bulk counterparts.*

In the initial experiments, a thin layer of 2D materials was deposited via spray printing, producing a noticeable colour change of the substrate. Nevertheless, SEM characterization revealed that full surface coverage was not achieved, with only a monolayer of 2D flakes present in areas where the ink was present. In later trials, a considerably thicker layer of selected materials was deposited by using a more concentrated ink and increasing the number of spraying passes. The assembly of the flakes under the deposition conditions was then assessed for each material, with a few examples are provided below.

Two-dimensional LaOBr samples produced via sonication-assisted exfoliation in IPA or in a mixture of water and IPA were first evaluated as a representative dielectric oxyhalides. Both AFM (**Figure 3**a,b) and TEM (**Figure 34c**) characterization revealed a wide dispersion in lateral sizes and thicknesses, with most flakes being around 10 nm thick and close to or more than 1 µm in width, thus exhibiting a very large aspect ratio (> 100). The bulk crystal exfoliates perpendicular to the c axis, forming well-faceted large flakes alongside many smaller fragments.

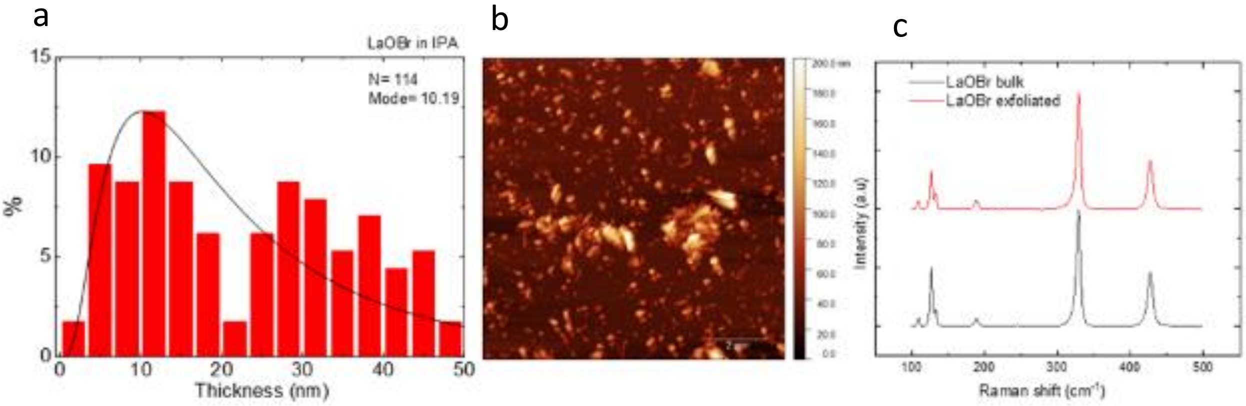


Figure 6. Characterization of LaOBr ink. AFM characterization of LaOBr ink with the thickness distribution (a) and an AFM image of flakes on Si wafter substrate. Raman spectroscopy characterization (c) reveals no notable difference between bulk and exfoliated material consistent with the thicknesses reported in (a)

In contrast, exfoliation in toluene produces flakes that are twice as thick. Future experiments will explore other exfoliating solvents to produce thinner flakes whilst maintaining their large lateral size. A representative film of spray coated LaOBr nanosheets is shown on Si wafer (**Figure 4**a) and on quartz (**Figure 4**b). Both bulk and the dispersion of 2D flakes in IPA are white in colour.

The spray coated LaOBr film features the platelets observed by TEM (**Figure 4c**) lying flat on the substrate or within the film in a closely packed arrangement. However, a considerable amount of volume is occupied by smaller fragmented flakes, some of which are randomly oriented (**Figure 4**d-f). This notably impacts on both porosity and surface roughness of the film. The impact of this arrangements on the performance of LaOBr as a 2D-printed gate dielectric is yet to be evaluated. Achieving a well-ordered film of closely packed 2D flakes will require reducing the polydispersity of the exfoliated and purified material, which will be tacked in future experiments.

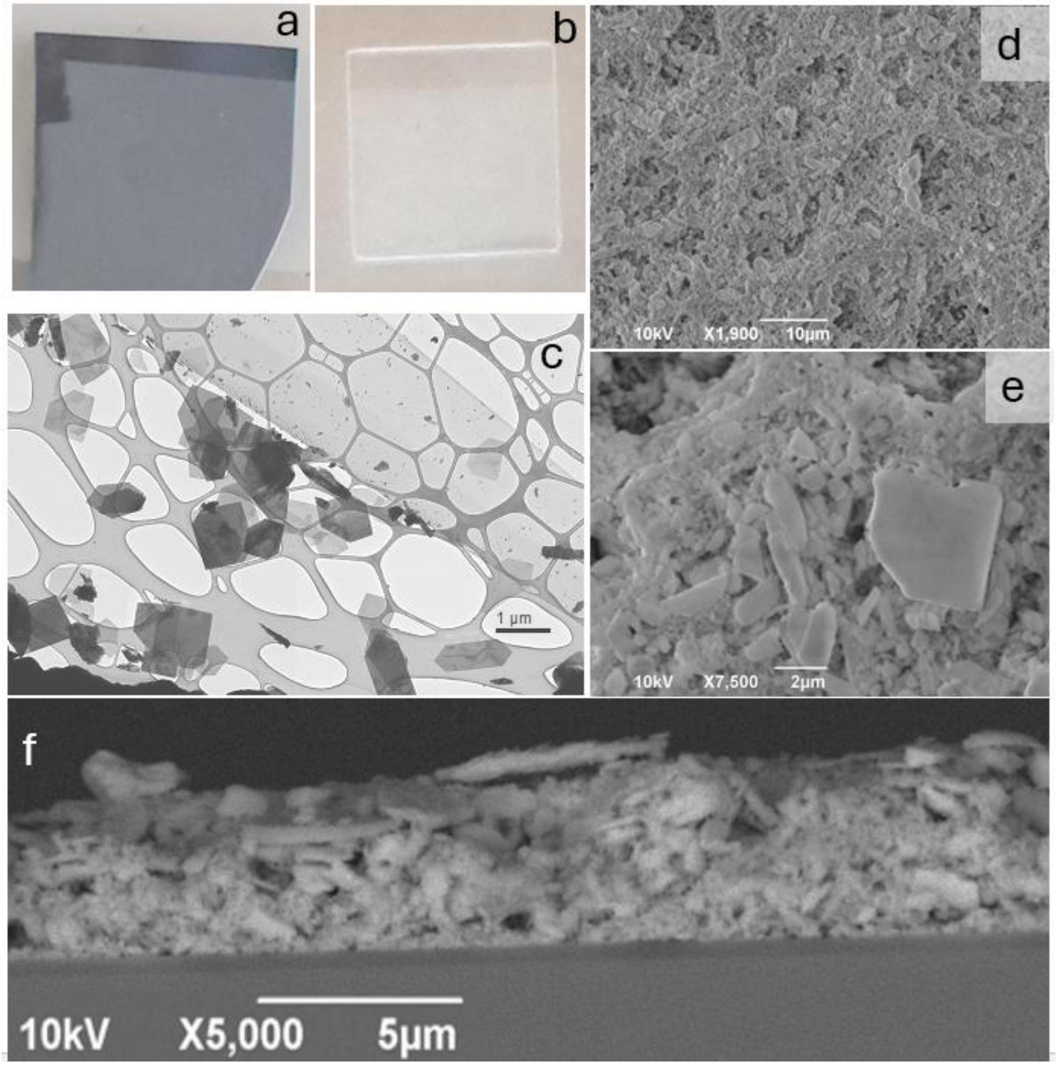


***Figure 7**. Spray printed LaOBr film. The LaOBr film on (a) Si wafter and (b) on quartz. TEM image of the ink (c) showing typical faceted platelets obtained by sonication assisted LEP in IPA. These platelets are also revealed in SEM images of the surface of the films taken a low (d) and especially at higher magnification (e). The cross section of the film deposited n SI wafer clearly shows many platelets layered on top of each other along with many smaller fragments oriented randomly.*

Figures 5 and **6** show the printed films produced from NdOBr and PrOBr inks, respectively. The sonication of bulk materials in IPA produces platelets with well-defined facets and narrower size distribution than LaOBr. The greater monodispersity of the flakes in the inks produced from these materials results in more compact films with uniform thickness and a smoother surface. In these films, densely packed nanosheets layered neatly on top of each other, result in a more cohesive structure. Furter optimisation of ink production and their complete characterization are ongoing.

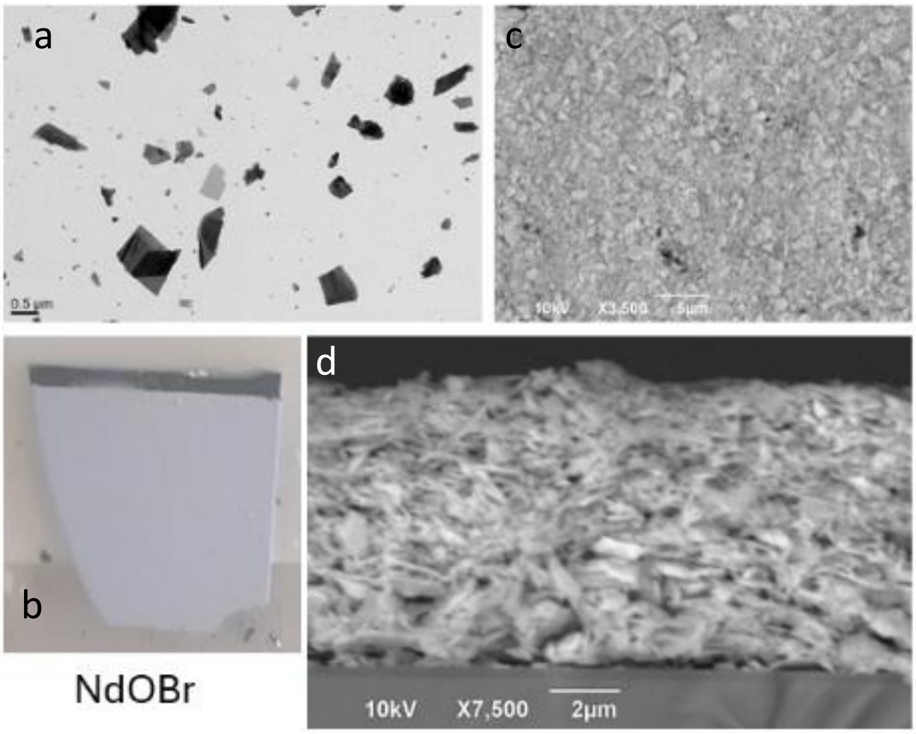


Figure 8. *Spray printed NdOBr film. Typical platelets obtained by LPE of NdOBr observed by TEM (a). Spray printed film of NdOBr ink on Si substrate (b). Low magnification (c) and cross section (d) SEM images of the spray printed film showing densely stacked 2D platelets forming a compact film.*

a

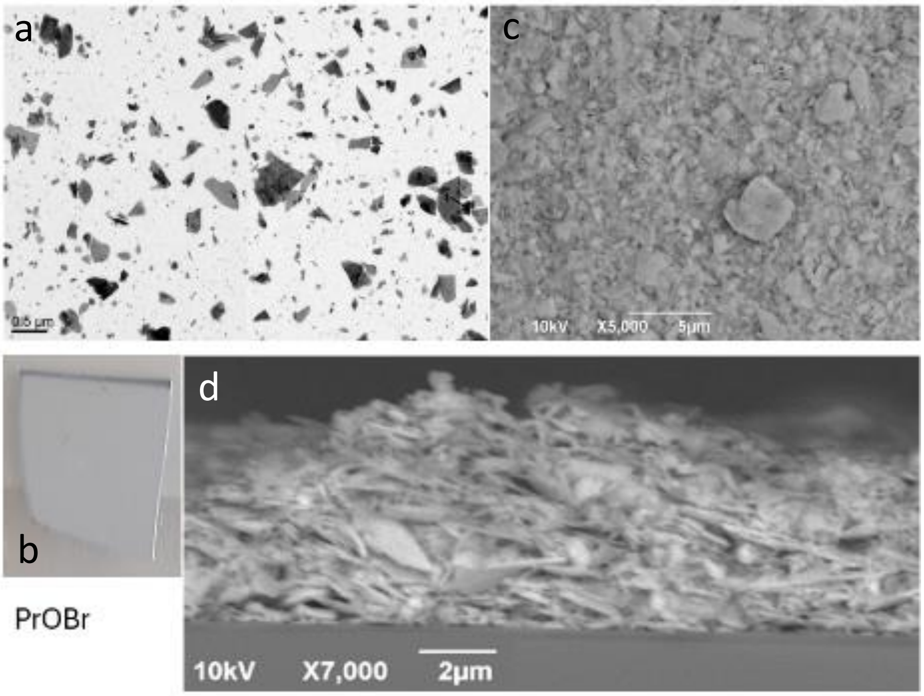


Figure 9*. Spray printed PrOBr film. Typical platelets obtained by LPE of PrOBr observed by TEM (a). Spray printed film of PrOBr ink on Si substrate (b). Low magnification (c) and cross section (d) SEM images of the spray printed film showing densely stacked 2D platelets forming a compact film.*

3.2 Contribution to project (linked) Objectives

These results significantly contribute to the overall project by addressing the challenges and optimizations required for novel 2D material ink formulations, stabilization, and deposition techniques. Here's a breakdown of how these findings advance the project:

1. **Ink stability improvements**: we show how specific solvents and stabilizing agents can prevent reaggregation and maintain the nanosheets' exfoliated state, ensuring the inks remain stable for various deposition methods. This allows for reliable, uniform coatings in processes like LS deposition. It sets the groundwork for utilizing novel 2D materials, such as dielectric oxyhalides, in similar high-throughput applications, enhancing the versatility of the project.
2. **Rheological optimization**: the precise control over ink viscosity ensures compatibility with different deposition techniques. This is critical for maintaining control over film morphology and improving the performance of devices fabricated using these inks. The detailed analysis of LS-deposited films, including graphene nanosheets, offers actionable insights into how concentration and mass loading affect the homogeneity and thickness of the final films. These optimizations directly impact the efficiency of deposition processes and minimize material waste.
3. **Exfoliation of novel materials**: the successful exfoliation and deposition of nine novel 2D materials, including dielectric oxyhalides like LaOBr, highlights the project's progress in expanding the range of usable 2D materials for digital electronics. The ability to control the thickness and lateral size of the flakes through different solvents or processing methods informs future efforts to optimize the quality and performance of these materials in various applications, such as printed electronics or dielectric layers.
4. **Deposition techniques**: the refinement of deposition protocols (e.g., spray-coating and LS deposition) for novel 2D material inks is crucial for creating uniform, high-quality and compact films. Our findings will guide future efforts to scale up production and improve the quality of deposited films across different materials and substrates.
5. **Characterization and data-driven insights**: the use of characterization techniques (e.g., SEM, TEM, AFM) to assess film quality, nanosheet dispersion, and homogeneity provides a detailed understanding of how material properties and deposition parameters affect film formation. This information will help in tuning the inks and deposition methods for specific applications, such as achieving smoother, less porous films for use in 2D-printed devices.

3.3 Contribution to major project exploitable result

The results of this deliverable contribute significantly to several major project exploitable results, particularly in terms of process optimization, product development, and scientific discovery. Here's how these contributions align with the project’s broader targets:

1. **Process optimization**: the refinement of ink formulations (through stabilization strategies using PVP for TMDs/TMMs) and the rheological optimization of the inks are key process to achieving high-throughput, scalable deposition methods such as spray-coating and Langmuir-Schaeffer (LS) deposition. These processes are crucial for enabling large-scale manufacturing of 2D material-based products, ensuring that the ink stability and deposition techniques can be effectively used in industrial settings. The optimization of LS deposition and spray-coating techniques for various 2D materials represents a major process innovation that will help streamline the production of thin films for a variety of applications (*e.g.*, printed electronics, sensors, coatings). The detailed insights into how ink concentration, nanosheet size, and solvent choice affect film quality will be critical for creating commercially viable processes.
2. **Product development**: the successful exfoliation and deposition of novel 2D materials like dielectric oxyhalides (e.g., LaOBr, NdOBr, PrOBr) contributes to the development of new 2D material-based products. These materials have potential applications in printed electronics, dielectric layers, and energy devices. For example, the discovery of optimal film thickness and uniformity could lead to high-performance 2D-printed gate dielectrics, enabling the next generation of flexible, thin-film transistors or other electronic devices.
3. **New design approaches**: our findings inform new design strategies for both the materials themselves and the deposition systems. For example, the understanding gained from LS-deposited films of graphene and other 2D materials can lead to the design of more efficient deposition systems or tailored ink formulations for specific substrates and application requirements. We therefore provide a framework for designing more uniform and homogeneous films, which are essential for reliable and reproducible performance in 2D material-based devices.

Overall, this deliverable contributes to the project's **exploitable results** by contributing to product development in printed electronics and coatings, and driving forward scientific discovery in the field of 2D materials. The methods and findings can be applied across industries, increasing the potential impact and commercialization of the project's outcomes

4 Conclusion and Recommendation

The results of this deliverable demonstrate significant advancements in the development and optimization of 2D material inks and their deposition processes. Through tailored stabilization strategies, rheological control, and careful selection of solvents, we successfully produced stable and scalable 2D inks, including graphene, transition metal chalcogenides (TMD/TMM), and novel dielectric oxyhalides. The refinement of deposition techniques, particularly the Langmuir-Schaeffer (LS) method and spray coating, allowed for improved film homogeneity and thickness control, laying the foundation for high-quality 2D material films for use in a variety of applications. We successfully demonstrated that the ink formulations can be tailored to meet the demands of different deposition techniques, ensuring compatibility with industrial-scale processes. The work also highlighted important scientific discoveries related to the behaviour of nanosheets during exfoliation and deposition, leading to better control over film morphology and material properties.

Recommendations:

1. **Further optimization of exfoliation techniques**: the sonication-assisted exfoliation of dielectric oxyhalides, such as LaOBr, yielded promising results but also highlighted issues with polydispersity in flake size. Future work should focus on optimizing exfoliation methods to produce thinner, more uniform flakes while maintaining large lateral sizes.
2. **Binder and additive usage**: future studies should explore the use of additives to enhance the mechanical properties and stability of the deposited films, especially for applications requiring higher film durability and robustness.
3. **Exploring additional solvent systems**: The results indicate that solvent choice plays a critical role in determining the final film properties. Further research should explore a wider range of environmentally friendly solvents to enhance both exfoliation efficiency and film deposition, particularly for novel 2D materials like oxyhalides.
4. **Scaling up deposition techniques**: to enable the commercialization of these inks and deposition methods, it is recommended to focus on scaling up the LS deposition at industrial level
5. **Application-specific testing**: the performance of the investigated 2D materials in specific applications, such as in printed electronics, sensors, and energy devices, should be further evaluated in subsequent 2D-PRINTABLE Work Package.

By continuing to build on the successes of this deliverable and implementing these recommendations, the project is well-positioned to achieve its goals of advancing 2D materials for scalable, industrial applications and contributing to the next generation of printed electronics and other technologies.

5 Risks and interconnections

5.1 Risks/problems encountered

If applicable (consider using table below to report risks – and solutions ! – encountered for the activities/tasks related to this deliverable)

Risk No.	What is the risk	Probability of risk occurrence[1]	Effect of risk[1]	Solutions to overcome the risk
WPx.x	Describe the risks here!! And please refer to the section of the text in the document dealing with this.	Indicate the level	Indicate the level	Give a description how to overcome the risk / describe give possible solution(s)

1) Probability risk will occur: 1 = high, 2 = medium, 3 = Low

5.2 Interconnections with other deliverables

If applicable

6 Deviations from Annex 1

If applicable

Report/summarise if/which deviations from the original plan have to be made

7 References

1. Lin Z*, et al.* Solution-processable 2D semiconductors for high-performance large-area electronics. *Nature* **562**, 254-258 (2018).

2. Backes C*, et al.* Production of Highly Monolayer Enriched Dispersions of Liquid-Exfoliated Nanosheets by Liquid Cascade Centrifugation. *ACS Nano* **10**, 1589-1601 (2016).

3. Carey T*, et al.* High-Mobility Flexible Transistors with Low- Temperature Solution-Processed Tungsten Dichalcogenides. *ACS Nano*, (2023).

4. Gabbett C*, et al.* Quantitative analysis of printed nanostructured networks using high-resolution 3D FIB-SEM nanotomography. *Nature Communications* **15**, (2024).

5. Kelly AG*, et al.* Highly Conductive Networks of Silver Nanosheets. *Small* **18**, (2022).

6. Wells RA*, et al.* High Performance Semiconducting Nanosheets via a Scalable Powder-Based Electrochemical Exfoliation Technique. *ACS Nano* **16**, 5719-5730 (2022).

7. Kelly AG, O'Suilleabhain D, Gabbett C, Coleman JN. The electrical conductivity of solution-processed nanosheet networks. *Nature Reviews Materials* **7**, 217-234 (2021).

8. Pinilla S, Coelho J, Li K, Liu J, Nicolosi V. Two-dimensional material inks. *Nature Reviews Materials* **7**, 717-735 (2022).

9. Backes C*, et al.* Spectroscopic metrics allow in situ measurement of mean size and thickness of liquid-exfoliated few-layer graphene nanosheets. *Nanoscale* **8**, 4311-4323 (2016).

8 Acknowledgement

The author(s) would like to thank the partners in the project for their valuable comments on previous drafts and for performing the review.

Project partners:

#	Partner short name	Partner Full Name
1	TCD	TCD THE PROVOST, FELLOWS, FOUNDATION SCHOLARS & THE OTHER MEMBERS OF BOARD, OF THE COLLEGE OF THE HOLY & UNDIVIDED TRINITY OF QUEEN ELIZABETH NEAR DUBLIN
2	UNISTRA	UNIVERSITE DE STRASBOURG
3	UKa	UNIVERSITAET KASSEL
4	BED	BEDIMENSIONAL SPA
5	TUD	TECHNISCHE UNIVERSITAET DRESDEN
6	VSCHT	VYSOKA SKOLA CHEMICKO-TECHNOLOGICKA V PRAZE
7	UNR	UNIRESEARCH BV
8	UniBw M	UNIVERSITAET DER BUNDESWEHR MUENCHEN
9	EPFL	ECOLE POLYTECHNIQUE FEDERALE DE LAUSANNE

Disclaimer/ Acknowledgment

This project has received funding from the European Union's Horizon Europe research and innovation programme under grant agreement No 101135196. Views and opinions expressed are however those of the author(s) only and do not necessarily reflect those of the European Union. Neither the European Union nor the granting authority can be held responsible for them.

9 Appendix A - Quality Assurance Review Form

The following questions should be answered by all reviewers (WP Leader, reviewer, Project Coordinator) as part of the Quality Assurance procedure. Questions answered with NO should be motivated. The deliverable author will update the draft based on the comments. When all reviewers have answered all questions with YES, only then can the Deliverable be submitted to the EC.

NOTE: This Quality Assurance form will be removed from Deliverables with dissemination level "Public" before publication.

Question	**WP Leader**	**Reviewer**	**Project Coordinator**
	NAME (Organisation)	NAME (Organisation)	Jonathan Coleman (TCD))
1. *Do you accept this Deliverable as it is?*	Yes / No (elaborate)	Yes / No (elaborate)	Yes / No (elaborate)
2. *Is the Deliverable complete?* - *All required chapters?* - *Use of relevant templates?*	Yes / No (elaborate)	Yes / No (elaborate)	Yes / No (elaborate)
3. *Does the Deliverable correspond to the DoA?* - *All relevant actions preformed and reported?*	Yes / No (elaborate)	Yes / No (elaborate)	Yes / No (elaborate)
4. *Is the Deliverable in line with the 2D-PRINTABLE objectives?* - *WP objectives* - *Task Objectives*	Yes / No (elaborate)	Yes / No (elaborate)	Yes / No (elaborate)
5. *Is the technical quality sufficient?* - *Inputs and assumptions correct/clear?* - *Data, calculations, and motivations correct/clear?* - *Outputs and conclusions correct/clear?*	Yes / No (elaborate)	Yes / No (elaborate)	Yes / No (elaborate)
6. *Is created and potential IP identified and are protection measures in place?*	Yes / No (elaborate)	Yes / No (elaborate)	Yes / No (elaborate)
7. *Is the Risk Procedure followed and reported?*	Yes / No (elaborate)	Yes / No (elaborate)	Yes / No (elaborate)
8. *Is the reporting quality sufficient?* - *Clear language* - *Clear argumentation* - *Consistency* - *Structure*	Yes / No (elaborate)	Yes / No (elaborate)	Yes / No (elaborate)

10 Appendix B