Data-Driven Smart Synthesis of Two-Dimensional Materials

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Introduction

Molybdenum disulfide (MoS_2) (Fig. 1a), a representative two-dimensional semiconductor, exhibits enormous potential in electronic and electrochemical applications [1]. While it can be synthesized using chemical vapor deposition (CVD), the process is far from optimization due to high-dimensionality of the synthesis parameter space and complex correlation among the process variables [2]. In this work, we exploited machine learning (ML) techniques to predict the success of MoS_2 synthesis and identified the crucial growth parameters. Leverage the knowledge gained, we successfully synthesized the material using CVD (Fig. 1b) and demonstrated high-performance MoS_2-based field-effect transistor (FET).

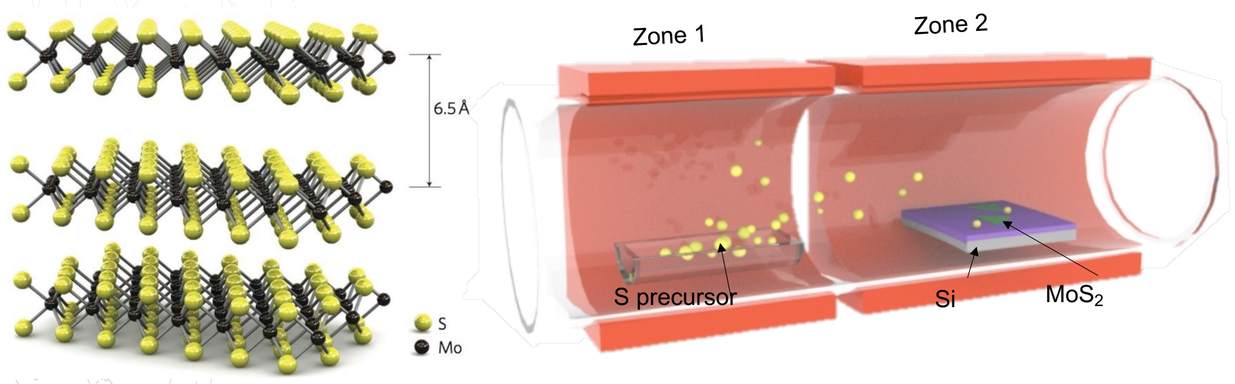


Fig. 1 Schematic illustration of (a) MoS_2 and (b) CVD.

Materials & Methods

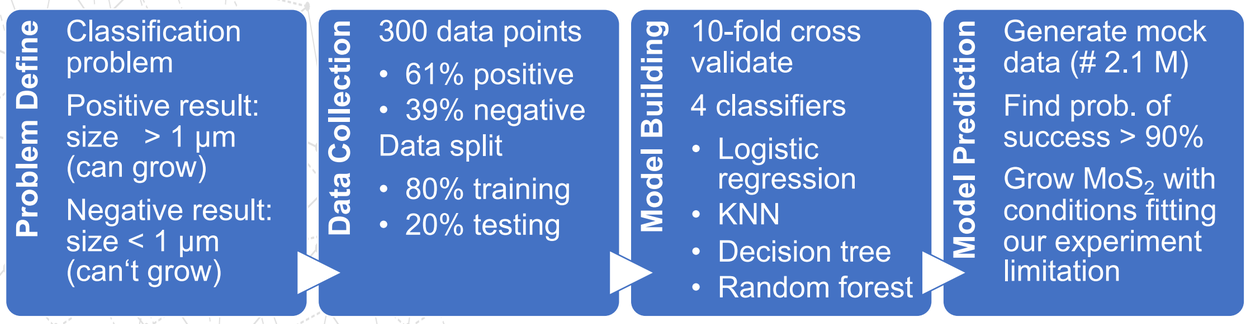


Fig. 2 ML model construction and analysis.

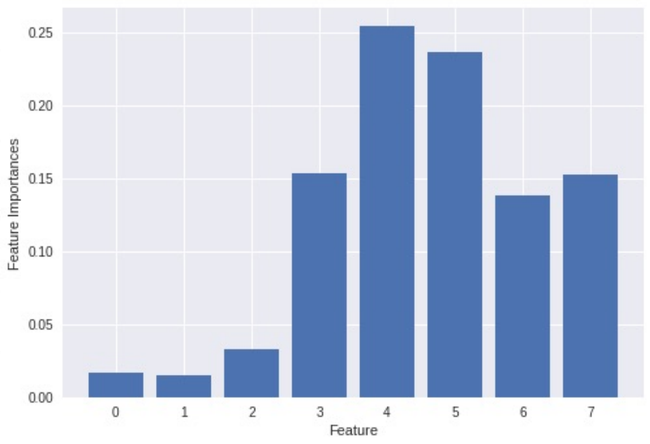
	Add NaCl	Dist. of S (in cm)	Flat/ Tilted	Flow rate (sccm)	Reaction temp. (°C)	Ramp time (min)	Reaction time (min)	Can grow
	0	2.0	Flat	50	500	13	10	0
	1	2.0	Tilted	200	550	20	5	1
	1	2.0	Flat	60	750	16	10	0
	⋮	⋮	⋮	⋮	⋮	⋮	⋮	⋮
Min	0	0.5	-	10	500	10	5	0
Max	1	3.5	-	250	975	30	15	1
Avg	0.3	2.1	-	72	728	16	12	0.6
Std	0.5	0.5	-	42	95	2	3	0.5

Table 1 Open source dataset for MoS_2 CVD growth [3].

Results

Model	Accuracy	Precision	Recall
Logistic regression	0.691	0.644	0.853
KNN	0.879	0.861	0.912
Decision tree	0.896	0.872	1.0
Random forest	0.913	0.868	0.971

Table 2 Performance comparison of ML classifiers.



The most significant factors affecting the success of MoS_2 CVD growth
- Gas flow rate (#4)
- Reaction temp. (#5)
- Reaction time (#7)

Fig. 3 Feature evaluation from random forest classifier.

Results cont.

Add NaCl	Dist. of S (in cm)	Flat/ Tilted	Flow rate (sccm)	Reaction temp. (°C)	Ramp time (min)	Reaction time (min)	Can grow pred.
0	2.9	Flat	80	750	19	14	1
0	1.7	Flat	80	800	23	12	1
0	1.7	Flat	75	780	39	22	1

Table 3 ML suggested growth conditions.

The ML proposes a set of growth condition with over 90% probability of success by exploring 2.1 millions of possible combinations.

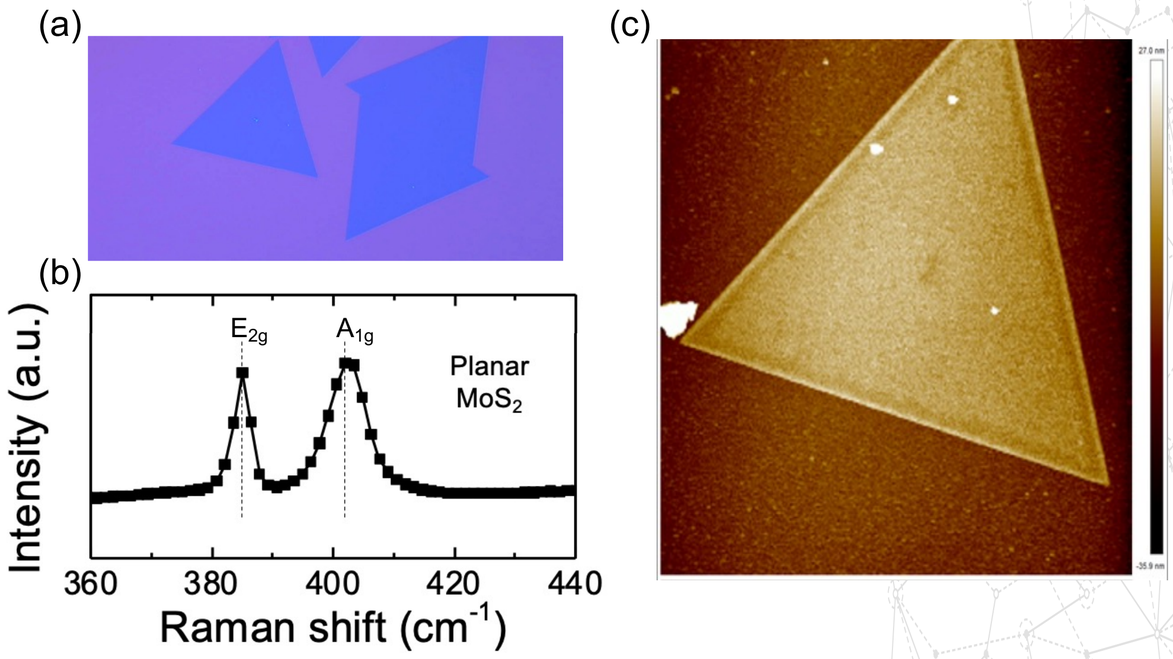


Fig. 4 (a) Optical image, (b) Raman spectrum, and (c) AFM image of as-synthesized MoS_2.

High crystallinity and monolayer MoS_2 was successfully synthesized using the ML-suggested optimal growth conditions.

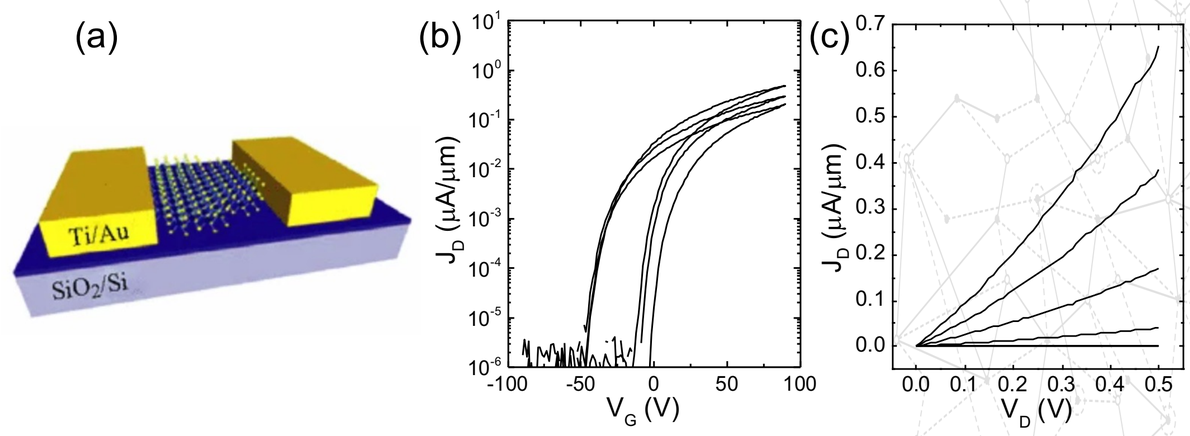


Fig. 5 (a) Schematic of MoS_2-based FET (b) Output and (c) Transfer characteristics of the device.

The MoS_2-based FET showed an on/off ratio of 10^7 and a typical n-type semiconductor behavior [4].

Conclusions

- The ML method can be employed to accelerate the exploration of CVD synthesis parameter space.
- The random forest model showed an 91.3% accuracy for the success of CVD synthesis prediction.
- The random forest model could improve the MoS_2 CVD experiments success rate from 61% to 90%.
- Monolayer MoS_2 with high crystallinity was successfully synthesized using the ML-predicted parameters.

Acknowledgement

This work is an MDI interconnect project (MDI 501 and 503, spring semester). Part of the experimental data is collected in the NETLab (Dr. F. Yao) with funding support from CMI under award C160186.

References

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