Self-Driving Labs for Nanomaterials Development for Energy Applications: Synthesis, Dispersion, and Composite Forming

Shun Muroga, Takashi Tsuji, Shunsuke Sakurai, Don N Futaba, Kenji Hata
National Institute of Advanced Industrial Science and Technology (AIST), Tsukuba central 5, 1-1-1, Higashi, Tsukuba, Ibaraki, 305-8565, Japan

Correspondence to: Shun Muroga, muroga-sh@aist.go.jp

1. Introduction

Nanomaterials exhibit outstanding functionalities owing to their high specific surface area. Among them, carbon nanotubes (CNTs) have attracted considerable attention because of their exceptional properties, and intensive research and development have been conducted. CNTs are reported to have approximately 20 times the strength of steel, about 10 times the thermal conductivity of copper, roughly half the density of aluminium, and about 10 times the electron mobility of silicon. Leveraging these characteristics, CNTs have been utilized in composites with polymers, rubbers, and metals, as well as in high-performance and energy-efficient applications such as battery and semiconductor-related materials, including those intended for extreme environments.

Meanwhile, the performance requirements for materials continue to increase, and identifying materials with desired properties from an enormous design space of material candidates and process conditions demands substantial time and effort. To address this challenge, we have been working on AI-driven analysis, including multimodal machine learning [1–3], self-driving laboratories (SDLs) [4], and R&D support using large language models. In this presentation, we introduce our SDL framework and analysis case studies aimed at capturing complex synthesis conditions and CNT behavior.

2. SDL of CNT Synthesis

Fig. 1 illustrates the concept of our self-driving laboratory for CNT synthesis (super-growth method [5]). In this process, CNTs grow vertically from catalyst nanoparticles on a substrate via chemical vapor deposition (CVD). The substrates are automatically transported by a robot and retrieved after synthesis, enabling efficient acquisition of data across a large number of conditions in a high-throughput manner.

Using the high-throughput CNT samples, we have also succeeded in building machine-learning models that relate synthesis conditions to CNT properties, providing guidance for condition optimization [6]. However, analyses based solely on final *ex-situ* properties have limitations in accurately capturing dynamic behaviors during synthesis, such as nucleation, growth, and catalyst deactivation. To overcome this, we performed *in-situ* measurements of CNT growth behavior, extracted features from the resulting time-series data, and integrated them into machine learning models to improve property prediction and to elucidate relationships among synthesis conditions.

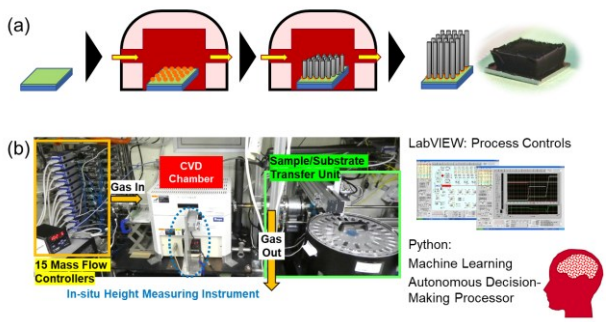

Fig. 1 (a) Schematic of synthesizing vertically aligned carbon nanotubes by chemical vapor deposition. (b) A photograph showing our developed self-driving laboratory of carbon nanotube synthesis.

3. Outcomes and Future Prospects

As a first step to developing our CNT-CVD SDL, we developed machine-learning models using data obtained both in situ and ex situ under various CNT synthesis conditions, and evaluated correlations among properties and predictive performance (Fig. 2). From the *in-situ* growth curves, we quantified the initial growth rate during the early stage of synthesis. We also defined the time scale from the early to the late stage of the process as the lifetime until catalyst deactivation and converted it into a numerical feature.

As *ex-situ* properties, we used yield, density, and the crystallinity indicator G/D ratio estimated from Raman spectroscopy. Together with the two in situ-derived variables, we set a total of five target variables. Gas flow rates were used as explanatory variables, and after comparing multiple modeling approaches, we report results obtained using Gaussian process regression (GPR) with a Matérn kernel.

Pair plots of the five CNT properties measured in this study showed the complex relationships among properties with some exhibiting trade-offs (Fig. 3(a)). Summarizing these modeling results showed relatively accurate prediction for the initial growth rate and G/D ratio but less so for lifetime and other variables (Fig. 3(b)). This suggests that gas mixing conditions may influence CNT nucleation and growth differently from how they affect catalyst deactivation, and such differences likely manifested as variations in predictive performance. With these results, we have a more well-defined search space and begin the introduction of orchestration to reduce the role of human in the loop.

Broadly, accelerating CNT synthesis via an SDL not only provides insights into conditions required to produce high-performance CNTs, but also offers a pathway to integrate with downstream SDLs for dispersion and processing. Such end-to-end linkage is expected to accelerate the development of high-performance functional materials and to help extract mechanistically relevant information for understanding complex, intertwined process-condition spaces.

References

[1] Muroga *et al.*, *Adv. Sci.*, 10, 24, 2302508 (2023).
[2] Kimura, Muroga *et al.*, *Carbon*, 241, 120390 (2025).
[3] Muroga *et al.*, *Appl. Spectrosc.*, 79, 1, 104-113 (2025).
[4] Muroga *et al.*, *Mater. Horiz.*, 12, 623-629 (2025).
[5] Hata, Futaba *et al.*, *Science*, 306, 1362-1364 (2004).
[6] Lin *et al.*, *ACS Nano*, 17, 22, 22821-22829 (2023).

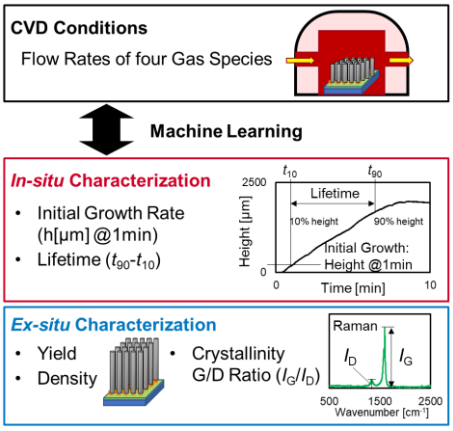

Fig. 2 Machine learning analysis of CVD conditions and in-situ and ex-situ characterization of CNT synthesis.

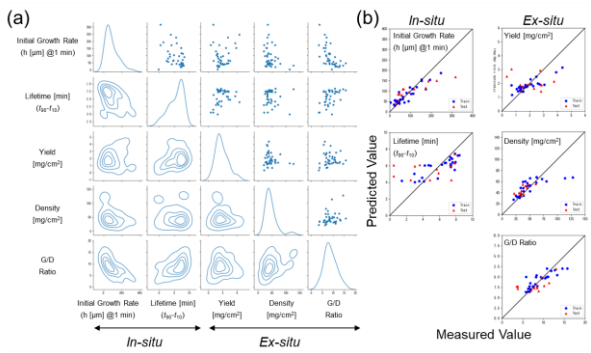

Fig. 3 (a) Scatter plot and kernel density estimation profiles of synthesized CNT properties. (b) Performance of machine learning model for predicting CNT properties obtained from *in-situ* and *ex-situ* characterization.