From synchrotron-dependency to in-house experiments: a self-driving lab for nanoparticle synthesis

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Self-driving laboratories (SDLs) are revolutionizing the throughput capabilities of lab-based chemical synthesis. Use of modern robotics is absolute key, as it allows for automated synthesis, i.e., efficient, reproducible, high-throughput chemical synthesis, whenever the synthesis recipe is at hand. The on-top addition of machine learning (ML) expands the capabilities to include autonomous synthesis, i.e., synthesis of target products without having the synthesis recipe at hand – but *learning* the synthesis recipe through efficient, explorative, data-driven synthesis screening [1]. Bayesian optimization (BO) algorithms have found profound use in ML problems for optimization of hyperparameter values. For SDLs, BO-based algorithms are used to tune the various variables at play during chemical synthesis, which otherwise make up an immensely complicated, multidimensional parameter space, e.g., for size and shape-specific metal nanoparticle synthesis [2].

High-throughput materials characterization of high fidelity calls for high-energy, high-intensity X-rays, i.e., X-rays from synchrotron sources. For nano materials, high-resolution powder X-ray diffraction (HR-PXRD) combined with Rietveld analysis falls short, due to pronounced broadening of Bragg peaks, resulting from the nanosized coherent domains, effectively making the information on the atomic structure unextractable. However, the information on the atomic structure becomes extractable, when using total X-ray scattering (TXS) combined with atomic pair distribution function (PDF) analysis [3]. In addition, small-angle X-ray scattering (SAXS) can provide complementary information on nanoparticle shape and size distribution, and X-ray absorption spectroscopy (XAS) can offer element-specific information on oxidation states (X-ray absorption near-edge spectroscopy, XANES) and local structure (extended X-ray absorption fine structure, EXAFS).

Unfortunately, synchrotron access is severely limited for most - if not all - academics and industry. Therefore, it is a *must* to abandon the otherwise high-fidelity synchrotron-based X-ray instrumentation and switch to lab-based X-ray instrumentation. The lower fidelity information from lab-based X-ray instrumentation must be compensated. Benefitting from the reproducibility of automated synthesis, when using robotics in the SDLs, this is done through parallel synthesis and complementary characterization; XTS for PDF analysis is complemented by SAXS as high-fidelity information sources, and lower-fidelity information is added through ultraviolet-visible (UV-VIS) spectroscopy. This constitutes the multimodal characterization loop of the in-house SDL for inorganic nanoparticle synthesis. All characterization methods add information, which the BO-based algorithm uses to learn the synthesis recipe, i.e., the optimal set of synthesis parameters for the target nano material in question.

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