Supporting Information for

Accelerated Screening of Colloidal Nanocrystals using Artificial Neural Network-Assisted Autonomous Flow Reactor Technology

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Experimental

Chemicals: Indium (III) chloride (99.999%), Zinc (II) chloride (> 98%), Oleylamine (OLAM) (technical grade, 70%), Octadecene (ODE) (technical grade, 90%), and tris-diethylaminophosphine (TDEAP) (97%) was purchased from Sigma-Aldrich. All solvents were degassed and stored inside the glovebox under nitrogen environment.

Indium Chloride in Oleylamine (0.3 M): 6 mmol indium chloride was added to a 50 mL three neck flask containing 20 mL degassed oleylamine inside the glovebox. The three-neck flask was then transferred to a Schlenk line for degassing (three cycles of vacuum and nitrogen purge) at 100 ºC for 1 hour. The stock solution was then kept under positive nitrogen pressure at 60 ºC under continuous stirring for flow synthesis experiments. Before starting the flow synthesis experiment, 24 mmol TDEAP was added to the flask and allowed to mix under continuous stirring.

Zinc Chloride in Oleylamine (1.0 M): zinc chloride stock solution was also prepared following similar step as indium chloride stock solution, except 50 mmol zinc chloride was added to 50 mL oleylamine inside the glovebox. The stock solution was kept under nitrogen at 60ºC with continuous stirring after degassing.

Automated flow synthesis of InP QDs: Indium chloride (with phosphorous precursor), zinc chloride stock solutions, and oleylamine solvents are pumped using three separate peristaltic pumps through an inline mixer (maintained at 60 ºC), followed by a nucleation and core growth reactor for synthesis of InP QDs. The ratio of zinc to indium precursors and the overall precursor concentration in the reaction mixture entering the nucleation-growth reactor are controlled by the flow rates of zinc chloride stock solution, indium chloride stock solution, and oleylamine solvent. The design of the automated continuous flow reactor platform used in this work is described in our previously reported work.[38,39] At different positions within the reactor channel, the reaction

mixture is sampled using a sampling valve (M-Switch from Fluigenet Inc.) and characterized using an inline UV-Vis and PL flow cell to monitor the reaction for different growth time. All process parameters including precursor flow rates, reactor temperatures, reaction sampling, inline characterization, and data analysis are controlled in an automated fashion using a Matlab script.

Ensemble neural network model: The ensemble model consists of 25 parallel artificial neural networks (as shown in Figure 3). For each of the network in this ensemble, the number of hidden layers is fixed as 2 The number of effective nodes in each hidden layer is 20 (half of the nodes in each layer is randomly dropped during the training, validation, and testing). A dropout layer with a value of 0.5 is used for all hidden layers in the ensemble network. A rectified linear unit is used as the activation function between each layer. For optimization, Adam algorithm with a learning rate between 0.01 and 0.001 is used to train the neural network. The median of output for each network in the ensemble is used to estimate the combined output and the standard deviation is used to estimate the uncertainty in predictions.

Supporting Figures

ENN-predicted features vs experimental data for testing sets as autonomous platform **performs** more experimental iterations:

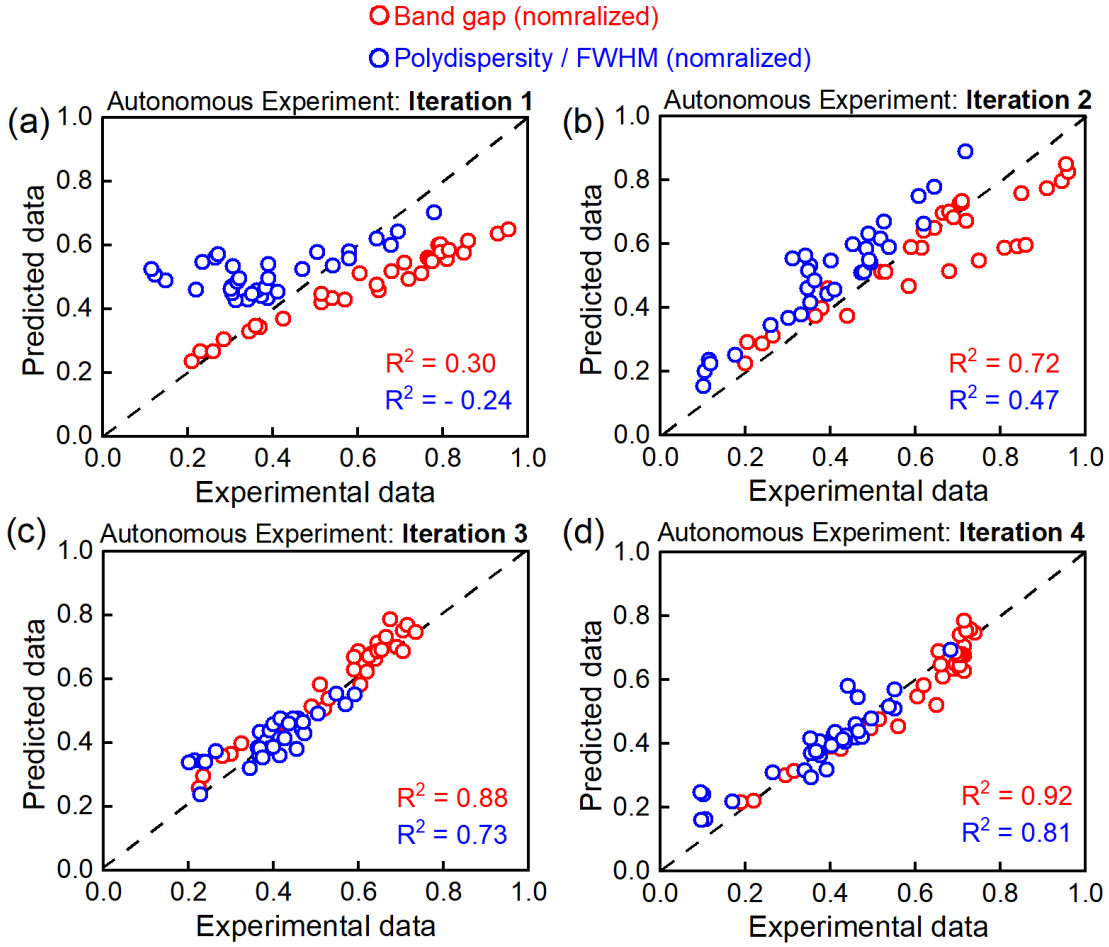

Figure S1. ENN-predicted band gap and polydispersity on testing data as the number of autonomous experiment iterations (training-validation-testing loop) increases. The accuracy of the model evaluated based on the testing data improves with progression of autonomous experimentation. The flowsheet of this iterative framework of model development using the autonomous experimentation platform is shown in Figure 1d

Relative frequency distribution of prediction certainty across the entire synthesis parameter space:

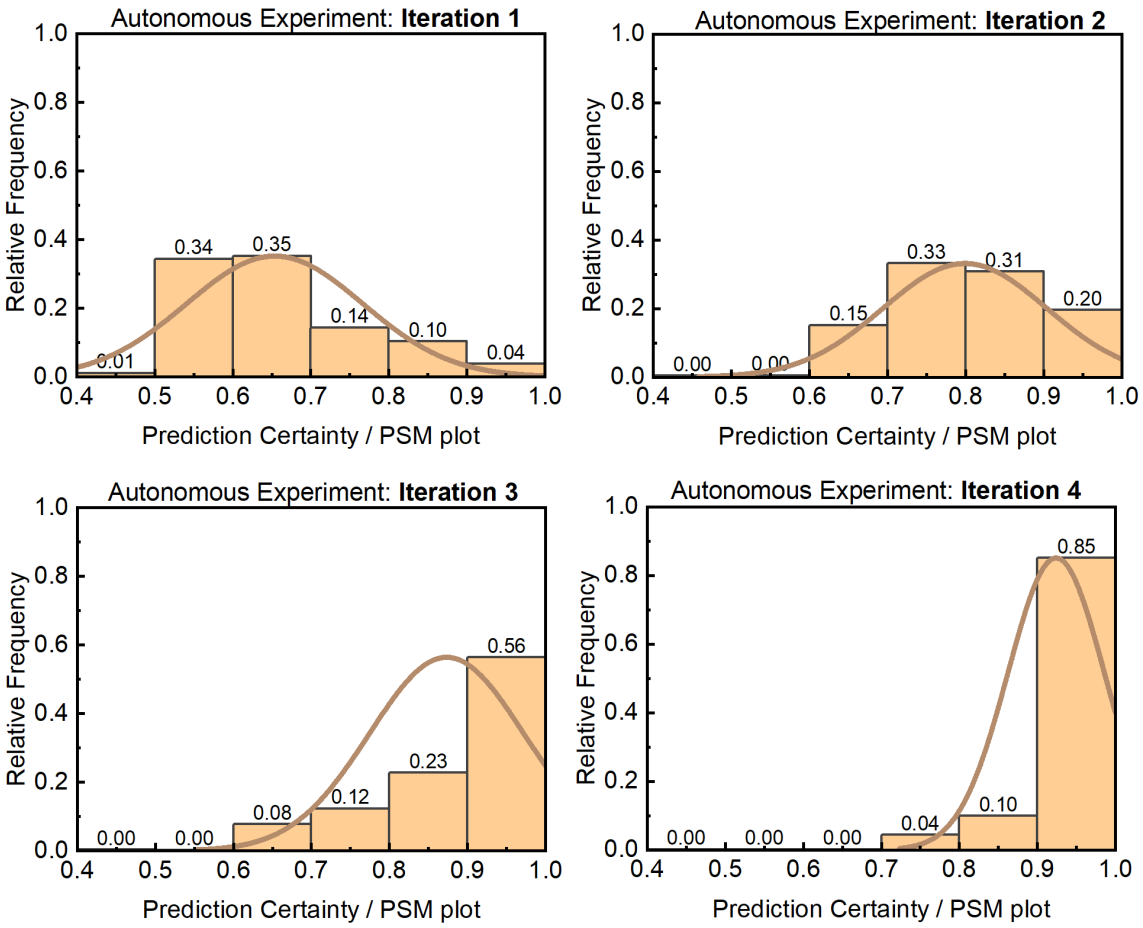

Figure S2. Relative frequency distribution of predictions with estimated certainty in prediction as the number of autonomous experiment iterations (training-validation-testing loop) increases. After four complete iterations of self-driven experiments, more than 85% of the entire synthesis parameter space is predicted with certainty higher than 0.90.

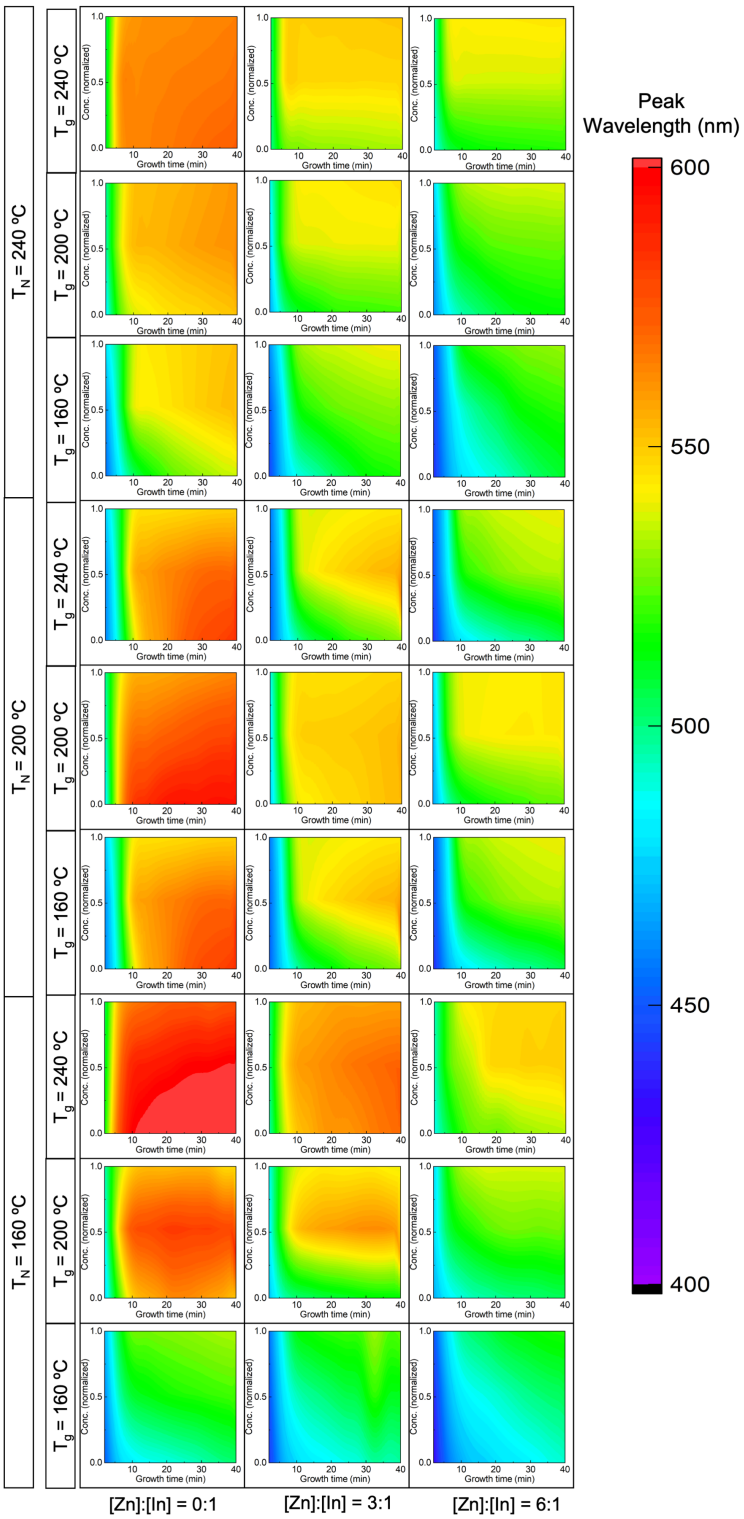

Figure S3. ENN-predicted peak wavelength (band gap) for different combinations of synthesis conditions.

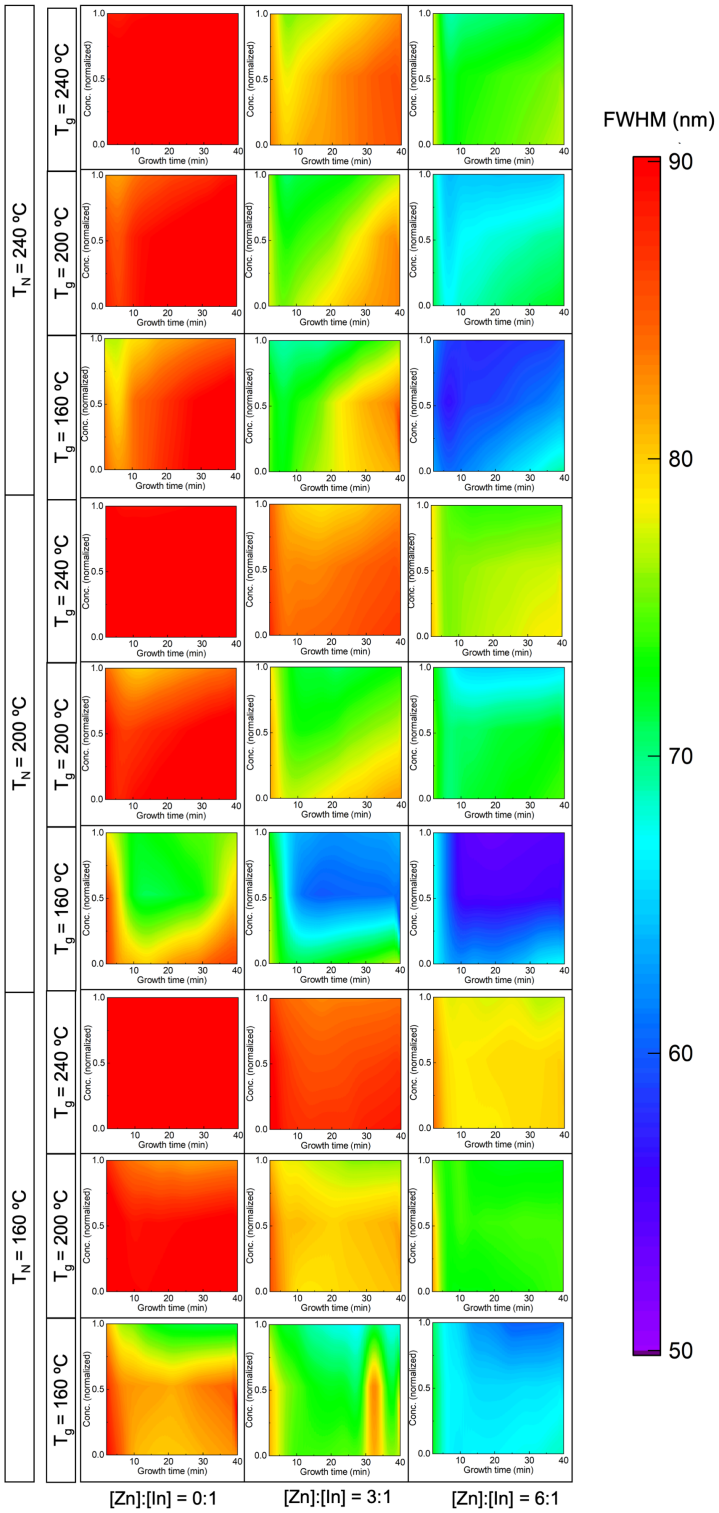

Figure S4. ENN-predicted FWHM (polydispersity) for different combinations of synthesis conditions.

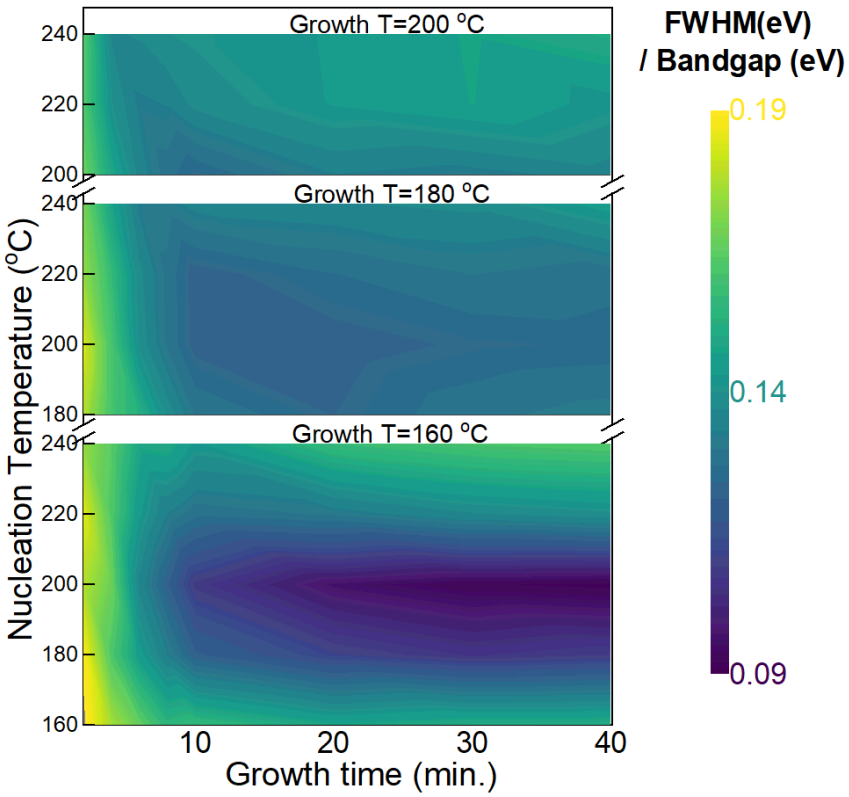

Figure S5. Polydispersity map expressed as FWHM (eV) / Bandgap (eV) fraction based on experimental data at different nucleation and growth temperature across the growth time (equivalent to the map shown in Figure 5d in the manuscript).