Neural network modeling and simulation of the synthesis of CuInS$_2$/ZnS quantum dots

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Abstract
The development of recipes for synthesis of quantum dots (QDs), a novel semiconductor material for application in optoelectronic devices, is currently purely based on experiments. Since the quality of QDs represented by quantum yield (QY) and emission peak strongly depends on a number of different parameters (route, precursors, conditions, etc), a large number of experiments is necessary. In this article, we show that data-driven modeling can be used as a supporting tool for optimization and a better understanding of the synthesis process. By using the results collected during the development of CuInS$_2$/ZnS (CIS/ZnS) QDs, a neural network model has been established. The model is able to predict the optical properties (QY and emission peak) of CIS/ZnS QDs as a function of the most important synthesis parameters, such as reaction temperature, time of CIS core formation and ZnS shell growth, feed molar ratio of Cu/In and Zn/Cu, various starting precursors, and types of ligands. Finally, a model analysis under various effects influencing the quality of QDs is performed.

KEYWORDS
CuInS$_2$/ZnS, neural network, optimization, quantum dots, simulation

1 | INTRODUCTION

During the past decade, semiconductor quantum dots (QDs) have been intensively studied due to their appealing optical properties such as high color purity, emission peak tunability, high brightness and photostability.$^1$ They have promising applications in solar cell, light emitting diodes (LEDs), sensors, fluorescent probes, biolabeling, and anticounterfeiting.$^2-12$ For example, QDs-based LEDs, so called QLEDs, exhibit longer photoluminescence (PL) lifetime and higher thermal stability than organic light emitting diodes$^{13}$ and, therefore, have been applied in the latest generation of displays.$^{14,15}$ However, currently used QLEDs, for example, in flat-screen televisions apply CdSe-based QDs. In spite of their excellent performance, concerns are growing due to the toxicity of Cd, especially when applying them in flexible/wearable displays. Therefore, CuInS$_2$ (CIS) QDs, considered as the alternative material, have attracted much attention because of their low toxicity, high optical absorption coefficients,$^{15}$ and high photochemical stability.$^{16-18}$

In above-mentioned applications, for example, in solar cells and LEDs, QDs with high photoluminescence quantum yield (PL QY) and in large range tunable emission wavelength are required. Core/shell QDs have the better chemical, thermal, and photochemical/physical stability compared with bare QDs.$^{19}$ Several approaches have been used to improve optical properties (PL QY and emission peak) of core/shell QDs, such as optimization of the interfacial alloyed shell...
thickness/structure compositions,20 selection of different shell materials,21 and process optimizations.22,23 In case of CIS QDs, ZnS is typically used as an inorganic shell to improve PLQY of QDs and adjust the emission wavelength.22,23 The optical properties of formed CuInS₂/ZnS (CIS/ZnS) QDs obviously rely on many parameters, such as reaction temperature and time of CIS core formation, ZnS shell growth temperature and time, feed molar ratio of Cu/In and Zn/Cu, various starting precursors, and types of ligands.24-30 These parameters can impact the synthesis and, in turn, optical properties of QDs. Due to the complexity of the synthesis of QDs, a large number of experiments is required in order to find an optimal recipe.

Given the current excitement surrounding artificial intelligence (AI), also chemical engineering is testing and partially rediscovering this approach. Novel developments in hardware and software allow to collect and process large amounts of data. Important component of the AI solutions is neural network (NN) modeling. In the published literatures, an NN is a powerful tool to handle nonlinear complex systems. It is comprehensively used in the system identification and control,31 quantum chemistry,32,33 decision making,34 pattern recognition,35 sequence recognition,36 medical diagnosis,37 financial applications,38 data mining,39 and visualization.40,41 In recent years, NN has been used as a powerful modeling tool in various chemical processes17,42-44 and in material sciences.45 For example, Hajjar et al17 used an NN model as a tool to predict the light olefins and gasoline-range selectivity and a genetic algorithm to optimize selectivity and operation conditions. In particular, NN models are well established for material development applications, for example, Sumpter et al45 elaborated an NN model for predicting the properties with different recipes and for designing materials that have desired physical and mechanical properties. However, until now, there are no trials to apply this approach for optimization of synthesis of new materials, especially nanoparticles. In the field of QDs synthesis, Singulani et al46 applied NN and genetic algorithms to predict the growth of self-assembled QDs. During the modeling, the indium flux in the reactor, the growth temperature, the deposition time, the width of the layer on top of which the dots are nucleated, and the aluminium and indium content in the InGaAlAs were selected as the input variables and the mean QD height was the output. The results showed that an NN is a good approximation of the function that governs the growth of QDs.

Against this background, an NN model might be to be a good supporting tool in order to rationalize the development process, get better understanding, and optimize reaction conditions. Certainly, there is burden caused by the tradition of the experimental approach. There are also rational reasons for the current status. From the data science point of view, even an extensive experimental investigation generates only a limited number of data. The recipes contain data of different type such as type of solvent or different synthesis steps that are not easy to replace with mechanistic descriptors. On the other hand, the combination of different factors might be critical for the final performance. Furthermore, it would be helpful to have a supporting tool that can streamline the development work for the synthesis with several operational steps and many parameters.

The work presented in this article aimed at validation if the model-supported development can be applied to the synthesis of new materials. In particular, we wanted to develop a model that will be able to predict the optical properties as a function of dossier for synthesis of CIS/ZnS QDs, and validate the NN modeling as the development tool to design synthesis of high performance QDs. As the database for the model development, a limited number of experiments well representing typical studies published in the literature has been used. In order to define the model, the parameters applied in the synthesis such as reaction temperature and time in CIS core formation, ZnS shell growth temperature and time, feed molar ratio of Cu/In and Zn/Cu, various starting precursors, and types of ligands have been selected as input variables and optical properties (PLQY and emission peak) have been set as output. The model has been quantitatively assessed by comparing model predictions with experimental data. Furthermore, the effect of important parameters has been simulated and discussed with respect to the physical interpretation of the predicted dependences. Finally, the model has been used for recipe optimization.

## 2 | MODEL AND EXPERIMENTAL DATA

### 2.1 | NN model

In many applications, an NN is a powerful tool to describe the input-output relationship of a technical system.47 We used the Bärmann software NN-Tool to generate the prediction model based on our experimental data. Thereby, a feedforward NN with one hidden layer is used, which in principal is able to approximate every given continuous function.48,49 Figure S1 shows the connection structure of the proposed NN model as presented in Data S1 part. The number of internal nodes in the hidden layer of an NN essentially determines the modeling ability of the system. The more internal nodes
are available, the more flexible is the network structure and the more complex relationships can be learned. However, with the increase of number of internal nodes, there is the danger of “overfitting,” that is, the network memorizes the data. In particular, the measurement errors will also be learned. The correct number of internal nodes is determined by the complexity of the functions to be learned, represented by the measured data. Therefore, the NNs are trained on the records of the training set for various network structures and their modeling accuracy is determined on the independent validation set. To reduce the dependence of the resulting network structure from the selection of a specific validation set, the method crossvalidation is used. Therefore, different validation sets are used such that each data point belongs exactly once to one validation set. The optimal network structure is chosen such that the mean value of the prediction error over all validation sets is minimal. Finally, the network with the optimal network structure will be trained on all records. In situations where the number of available experimental data is rather small, crossvalidation has the advantage that the final model is trained with all data and thereby avoiding overfitting. The main advantage of the used software tool is that the underlying training algorithm is extremely efficient. This efficient algorithm enables the use of crossvalidation and the adaption of the network complexity to the given data, thereby keeping the computing time maintainable.

2.2 Database

Altogether, a set of 94 experimental results has been used in this study. All the experimental conditions and obtained results are presented in the SI part (see Table S1). Detailed information on experimental, equipment, procedures, and analytics is given in the SI as well as in References 23, 53 and 54. From the full database, 80% of the experimental data were used for the training set and the other 20% of the experimental data were used as validation set, respectively (5-fold crossvalidation). Hence, each experimental data point belongs exactly once to one of the validation sets.

The main operating variables affect the optical properties of QDs very much; therefore, the effects of reaction time and temperature in CIS core formation, shell growth time and temperature, Zn/Cu feed molar ratio, and types of ligands are observed. Detailed operational or simulated conditions in investigation are listed in Table 1. According to the stoichiometry, the Cu/In feed molar ratio was fixed to one. The reaction time and temperature during core synthesis, shell growth time, shell growth temperature, Zn starting materials, ligands, and Zn/Cu feed molar ratio were selected as input variables. We determined the boundary values of these parameters as follows: CIS core reaction time from 30 to 120 minutes; CIS core reaction temperature at 240°C and 260°C; DDT (43 experiments), and TOP (eight experiments) as ligands; Zn(Ac)₂ (71 experiments) and ZDC (16 experiments) as Zn starting materials; shell growth time from 0 to 180 minutes; shell growth temperature from 200°C to 240°C; and Zn/Cu feed molar ratio from 1 to 8. CIS/ZnS QDs with PL QYs from 1.4% to 80.5% and emission peaks from 558 to 684 nm were obtained. An NN has been trained based on these data. The input layer of the network contains one neuron, and the output layer contains two neurons. For both output parameters, respectively, eight knots were used in the inner layer, resulting in a prediction model with optimal validation set behavior (crossvalidation).

3 RESULTS AND DISCUSSION

3.1 Validation of the NN model

In the first step, the quality of the model of the core, that is, CIS QDs has been analyzed. The parity plots for the wavelength and the value of the QY peak are presented in Figure 1. The accuracy of the prediction expressed by the coefficient of determination \( R^2 \) amounts to 99.8% and 87.7% for PL emission peak and QY, respectively. Since the linear correlations are almost congruent with the diagonal of the parity plot, it might be concluded that the model is not biased by a systematic error, that is, the model covers all important effects. In spite that the database for the synthesis of pure CIS particles is quite limited, the proposed model comprises a quite wide range of the wavelengths and quantum yield peaks.

In the second step, the model has been extended on the synthesis of the core/shell particles, that is, CIS/ZnS QDs. The parity plots for PL emission peak and QY are shown in Figure 2. The coefficients of determination \( R^2 \) that amount to 95.1% and 89.8% for the both parameters, respectively, are very high when considering that the database comprises experiments in which seven inputs were varied. The experimental database covers a wide range of wavelength from 550
to 690 nm and QY up to ca. 80%. Again, no systematic error could be identified between experimental data and predicted values, for example, only a small deviation of the linear correlation of the data from the diagonal. Obviously, the model deals well with the complexity of the problem expressed by the large number of variables and nonlinear dependences. Finally, the limited accuracy of the experimental data has to be considered.

The previously aggregated analysis confirmed good statistical accuracy of the model. In the following, it will be shown that the model is able to reproduce physically relevant dependences. As an example, the effect of the variation of the reaction time during synthesis of the core CIS particles on PL emission peak and QY was simulated (see Figure 3). This effect has been already studied, also by other groups. Figure 3 illustrates a very good agreement between the model predictions and the experimental data. Furthermore, the model reproduced well the observed nonlinear dependences that have physical background. The observed redshift of emission peak can be explained by the reduced confinement effect...
related to the growth of particles and the evolution of QYs is related to the numbers of surface defects. For the detailed discussion, we refer to our previous article. 54

3.2 | Simulations

In this paragraph, simulation results obtained using the developed NN model will be presented. The study aimed as deconvolution of different effects and illustration that the model predictions could be well interpreted with respect to the physics of PL.

Effect of time of the core synthesis on optical properties of CIS/ZnS QDs. It can be seen from Figure 4 that evolution of PL emission peak and QY of CIS/ZnS QDs exhibits the same tendency as it has been observed for the synthesis of the core CIS. It might be concluded that there is a strong correlation between optical properties of CIS core and CIS/ZnS QDs. However, the ZnS shell shifts the peak to lower wavelengths, in average ca. 50 nm. The effect of the shell is especially distinct for small cores, that is, for CIS core synthesized at times below 90 minutes. Figure 4B shows that PL QYs of CIS/ZnS QDs always exhibits a maximum which is significantly higher than the one of the CIS cores. However, there is a correlation between maxima of CIS and CIS/ZnS QDs. In conclusion, QYs of CIS core is critical in order to achieve high-performance QDs. Obviously, the performance of the CIS/ZnS QDs is strongly influenced by the synthesis shell and, in particular, thickness of the shell. There is a general agreement that growth of ZnS shell reduces surface defects of nanocrystals and therefore improves optical performance of CIS/ZnS QDs. Highest QYs of CIS/ZnS QDs have been achieved for shells synthesized between 60 and

**FIGURE 4** Model predictions of the effect of core reaction time and shell growth time to (A) PL emission peak and (B) QY of CIS/ZnS QDs. Simulation condition: Ligand = DDT, Zn-starting material = Zn(OAc)₂, core reaction temperature = 240°C, Zn/Cu feed molar ratio = 4, and shell growth time from 0 to 180 minutes

**FIGURE 5** Model predictions of the effect of shell growth time and temperature to (A) PL emission peak and (B) QY of CIS/ZnS QDs. Simulation condition: Ligand = DDT, Zn-starting material = Zn(OAc)₂, core reaction temperature = 240°C, core reaction time = 60 minutes, Zn/Cu feed molar ratio = 4, shell growth temperature = 240°C, and shell growth temperature from 200°C to 240°C
150 minutes. Obviously, too thin and too thick shells are suppressing high QYs and the model enables to identify optimal parameters.

**Effect of shell growth time and temperature on optical properties of CIS/ZnS QDs.** As illustrated in Figure 5A, growth of ZnS shell causes the spectral blueshift. Moreover, the evolutions of PL emission peak of QDs always present a dramatical blueshift at the beginning and then followed by slight reduction of the wavelength with the further growth of ZnS shell. The dramatic blueshift at the beginning is caused mainly from the reduction of surface defects, while the thickness of ZnS shell has no obvious contribution. During the growth of ZnS shell, the ion exchange of core and shell induced the shrink of CIS core, which lead to the continuous blueshift and even became dominated at latter time. The detailed mechanistic interpretation is given elsewhere.54 Higher shell growth temperature can accelerate the shrink of CIS core and induce larger blueshift. Other claims regarding the mechanism of blueshift are CIS surface reconstruction,56 etching of the plain core materials,24 or the interdiffusion of CIS and ZnS57,58 or alloyed interfacial layer formation during shell growth.28 The dependence predicted for the CIS/ZnS QDs differs to the small redshift reported for type I core/shell heteronanocrystals, which is caused by the leakage of the exciton wave function into the shell.59,60 The blueshift comes with exciton-exciton repulsion and redshift from their attraction.61

Figure 5B presents the evolution of PL QY of CIS/ZnS QDs due to the growth of ZnS shell. In addition, this dependence exhibits a maximum. The reasons for the QY growth below the maximum have been already discussed. The decrease of the yield above the maximum is caused by the surface defects that are generated due to the decomposition of DDT.54 The same behavior has been predicted in the whole temperature range (220°C-240°C). The maximum PL QYs can typically be obtained at the shell growth time ranging from 40 to 100 minutes. Regarding the effect of shell growth temperature, a high temperature improves PL QYs of QDs. According to the model predictions through optimization of shell growth time and temperature, predicted maximum QY of CIS/ZnS can reach up to 80%. The simulated dependencies and predicted optical performance are consistent with results of experimental studies.23

### 3.2.1 Effect of ligand type on optical properties of CIS/ZnS QDs

The type of ligand is one of the key factors that influence the optical properties of CIS/ZnS QDs when using DDT or TOP as the ligand (see Figure 6). It is obvious that the ligand type has critical impact on the optical performance but the effect of other variables, for example, the time of the shell growth is quite similar. The simulation results are conformed with our previous research.53,54 TOP can coordinate with ZDC to form complex and as weak Lewis base efficiently interact with weak Lewis acid (Cu⁺) at low temperature, this effect greatly increases the precursor activity, resulting in faster growth rates of the ZnS shell. During the growth of ZnS shell, the interaction ability of DDT and particles is stronger compared with TOP. Therefore, DDT can effectively reduce the surface defects and improve emission efficiency significantly. In case of TOP, two factors play a role: TOP can reduce the surface defect as the organic layer; however, it also represents as a hole receptor inducing the probability of nonradiative relaxation.53,62,63

![Figure 6](image-url)  
**Figure 6** Model predictions of the effect of types of ligands to (A) PL emission peak and (B) QY of CIS/ZnS QDs. Simulation condition: Zn-starting material = ZDC, core reaction temperature = 240°C, core reaction time = 60 minutes, shell growth temperature = 200°C, and Zn/Cu feed molar ratio = 4.
3.2.2 | Effect of reaction temperature during core synthesis on optical properties of CIS/ZnS QDs

The rise of the temperature during core synthesis has significant impact on the optical properties (see Figure 7), for example, when rising the temperature only by 20 °C from 240 °C to 260 °C, the yield decreased from almost 80% to ca 10%. Simultaneously the wavelength increased by ca. 70 nm. The same behavior has been predicted for all times of the growth of shell, that is, thicknesses of the shell. The dramatic decrease PL QY with the increase of reaction temperature can be explained by the large amounts of defects formed in the core particles due to fast growth of crystal. Interesting is that these defects could not be repaired by the shell even if the shell growth was long. With respect to the shift of the wavelength, it is in line with previous discussion. Typically, increase of temperature can accelerate the growth of particles, in turn, larger emission wavelength generated due to quantum confinement.

In conclusion, the analysis on the effect of typical parameters to optical properties of CIS and CIS/ZnS QDs based on the model is fully conform with our former experimental results, showing that the NN model can be used as a development tool to well predict the optical properties of QDs and design synthesis of the high performance QDs.

3.3 | Optimization

The general purpose of the development of the model is to design a recipe that yield QDs with a high yield and controllable wavelength. The model can help in two ways. The first one is the learning effect. Better understanding of the governing mechanisms allows to make rational decisions. An experimental optimization is also most effective if the critical parameters are known. With this purpose, that is, identification of the most influencing factors, a sensitivity analysis has been performed. The second way is through pure mathematical optimization. This approach is the most effective. However, the NN models are only valid in the range of conditions applied in the experimental studies. If the optimum is outside of this range, an iterative approach of simulation and experiments has to be applied.

3.3.1 | Sensitivity analysis

As discussed, many parameters will influence the optical properties of CIS/ZnS QDs, but some key parameters have main contributions and meanwhile these parameters are not always same effect tendency, that is, they can positively
or negatively influence the QY. With the help of sensitivity analysis, the influence of the different input parameters to
the output parameters could be ranked as is shown in Figure 8. The average value of the derivative of the output with
respect to an input over all data points is calculated and compared for the different input parameters. The priority of
the variables that influence PL QY as shown in Figure 8A is core reaction temperature > shell growth temperature > core
reaction time > types of ligand > Zn/Cu feed molar ratio > shell growth time > types of starting materials. The result of
this formal analysis is in line with the expectation since for the high PL QY, well-organized crystal structure is necessary.
Simplifying, it is better not to make defects than to repair them effectively. The priority of the variables that influence
the wavelength of the PL emission peak is as follows (see Figure 8B): shell growth time > CIS core reaction tempera-
ture > types of ligands > CIS core reaction time > Zn/Cu feed molar ratio > shell growth temperature > types of starting
materials. However, all results presented in the previous section indicate that the wavelength of the peak and the PL QY are
correlated.

3.3.2 Final optimization result

During a model-based optimization, a combination of a Monte Carlo method followed by a gradient-based local optimi-
zation is used. Hereby, the best results of the Monte Carlo method are used as initial points for the gradient based
optimization. As high PL QY with defined emission wavelength is expected and interested in applications, the different
operational condition is simulated and optimized at different emission peaks. The peaks at around 550, 600, 650, and
700 nm were selected as targeted emission wavelengths. The reaction condition has been optimized to achieve high QYs
at each targeted wavelength. The optimization indicates that the QY of 90% should be possible. However, this point is
outside of the validity of the model. Therefore, it will be used in future studies for setting the strategy in the next wave of
experiments, that is, fixing DDT as ligands and CIS core reaction temperature at 240°C, the QYs with emission peak at
around 600 and 650 nm can reach up to 90% through optimization of other parameters. The detailed information of the
optimization studies is summarized in the SI part (Table S2).

4 CONCLUSIONS

In total, 94 sets of experiment were conducted, and the effects of the input parameters on the optical properties of CIS and
CIS/ZnS QDs were investigated. The optical properties of QDs highly depend on the reaction parameters. An NN tool was
presented to predict the optical properties of samples in the presence of seven input variables, in this study. The scatter plot
figures showed that the proposed model can well predict the optical properties of CIS and CIS/ZnS QDs with a correlation
coefficient value above 90% between experimental results and predicted values. The effect of several parameters in the
simulation is in accordance with our experimental results presented in previous publications. A combination of a Monte
Carlo method followed by a gradient-based local optimization was employed to obtain the optimal optical properties of
CIS/ZnS QDs and the appropriate reaction conditions. The present NN model can be used as a development tool to design
synthesis of high-performance QDs. However, it is more reliable for prediction within the tested regions. As is the case for any purely data-based models, it is not capable to extrapolate outside of this range. If a wider region is of interest, further experiments with parameters covering these regions should be conducted to verify the model. The wide experimental dataset is available in the supporting information and could be used by other researchers to extend it and develop a global model.

In general, the development of new materials is experimentally driven and based on the physicochemical interpretation of the results. This phenomenological approach is the only option since, due to the complexity of the problem, the development of a mechanistic model is not a viable option. The quality of the performance materials depends on the precursors, synthesis routes, conditions, and so on. Some of this information is reported as unstructured data. As a consequence, knowledge is represented by know-how and understanding rather than quantitative models. In other disciplines that cope with similar problems, significant progress in application of AI is observed. In this article, we tested if data-driven modeling can be applied in the optimization of the recipe for synthesis of QDs, which is an example of problems occurring when developing performance materials. The question occurred if the methods that have been successfully applied to "big data" problems could be also applied for the case that the number of data is limited compared with the input variables. The question has been certainly positively answered. Even a retrospective analysis as performed in this study brings a benefit, that is, a quantitative model that allows deconvoluting the effect of various influences in the process synthesis. However, we see the largest value in the parallel development and use of the model as a supporting tool in the design of the experiments and dynamic optimization.

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CONFLICT OF INTEREST
The authors declare no conflict of interest.

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