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USE OF NEURAL NETWORK APPROXIMATION OF THE PARAMETERS-PROPERTY RELATIONSHIP IN SYNTHESIS OF CARBON DOTS *

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Properties of Carbon Dots (CD)

CDs are nanoparticles with very interesting and useful properties

- Unique optical properties
    - Intense and stable photoluminescence (PL)
    - PL depends on wavelength of exciting radiation
- Easy to obtain in laboratory conditions
    - May be doped with various heteroatoms
    - Surface may be easily functionalized to modify properties
- Unique biological properties
    - Non-toxic
    - Bio-compatible
- Wide areas of use:
  optoelectronics, biomedicine, sensors, analytical chemistry etc.

Hydrothermal Synthesis of CD

Based on heating of solutions of carbon-containing material in a closed volume

- Parameters of synthesis that affect CD PL properties
  - Synthesis conditions
  - Sets of precursors
  - Ratio of their concentrations
- CD properties that are affected
  - Wavelength of PL radiation
  - Quantum yield of luminescence (QYL)
  - Photostability

Synthesis – Parameters – Properties

- A subset of widely known set of problems solved by modeling:

  Quantitative Structure – Property Relationships (QSPR)

- QSPR models predict properties of new substances/compounds

- Here we consider creation of an inverse QSPR model using machine learning (neural networks)
- Such type of model determines optimal parameters of synthesis allowing one to obtain substances with required properties
- We build a model to determine optimal parameters of synthesis to obtain CDs with required value of PL quantum yield

Materials and Methods

Hydrothermal synthesis of CD samples from the following precursors:

- Citric acid (CA) at fixed concentration (0.1 M)
- Ethylenediamine (EDA) in the range 0.01 M – 2 M

Synthesis conditions:

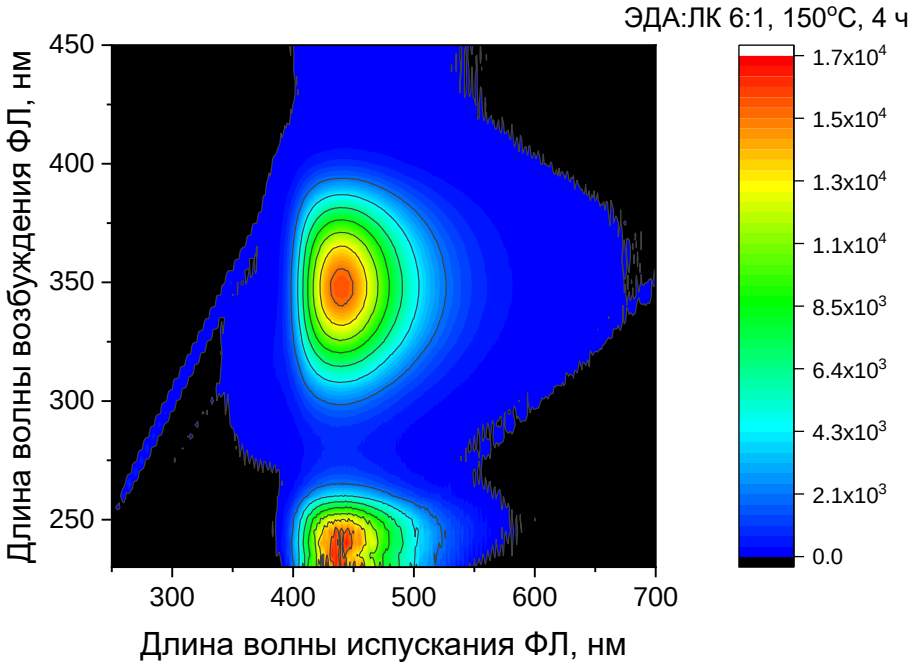
- Reaction time from 30 to 360 minutes
- Temperature of synthesis 80 to 200 °C

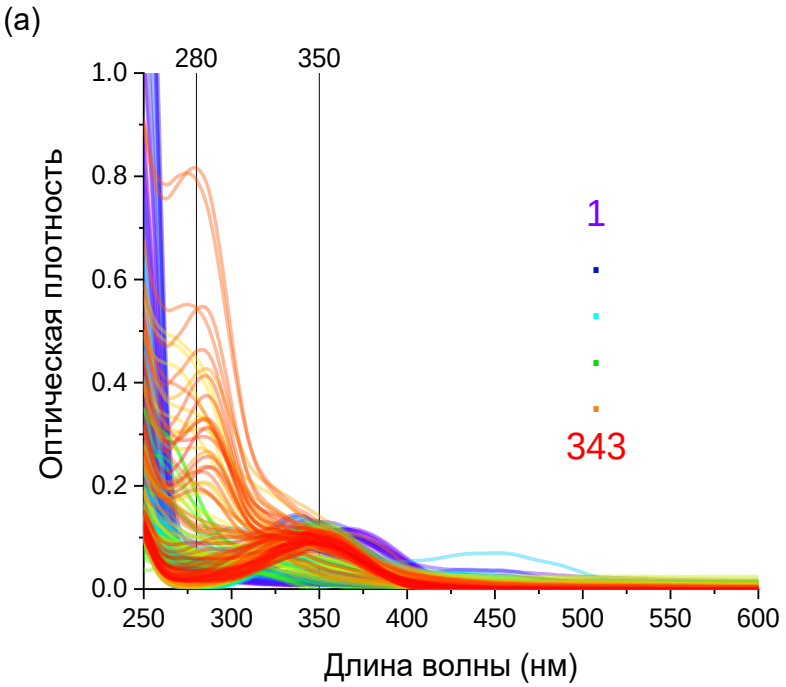
Total 343 CD samples were obtained. For each sample:

- Optical absorption spectrum obtained
- 2D fluorescence spectrum obtained
- QYL value calculated by the method of reference dye

Purpose: determine the area of the parameters
that allow one to obtain CD with highest QYL

Characteristic Spectra





Matrix of excitation/emission (2D fluorescence spectrum) of an aqueous suspension of a CD sample.

Optical absorption spectra of aqueous suspensions of CD samples.

QYL values were calculated from such spectra.

Synthesized Samples

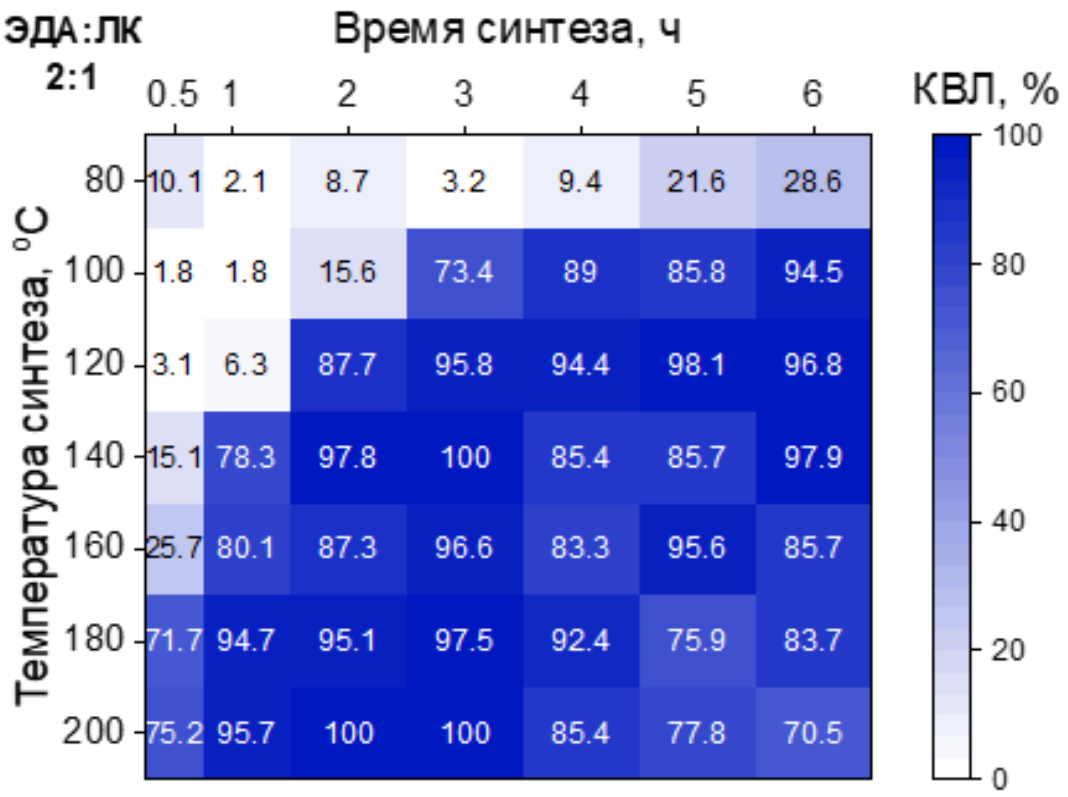


Photo (left) and QYL table (right) of synthesized samples.

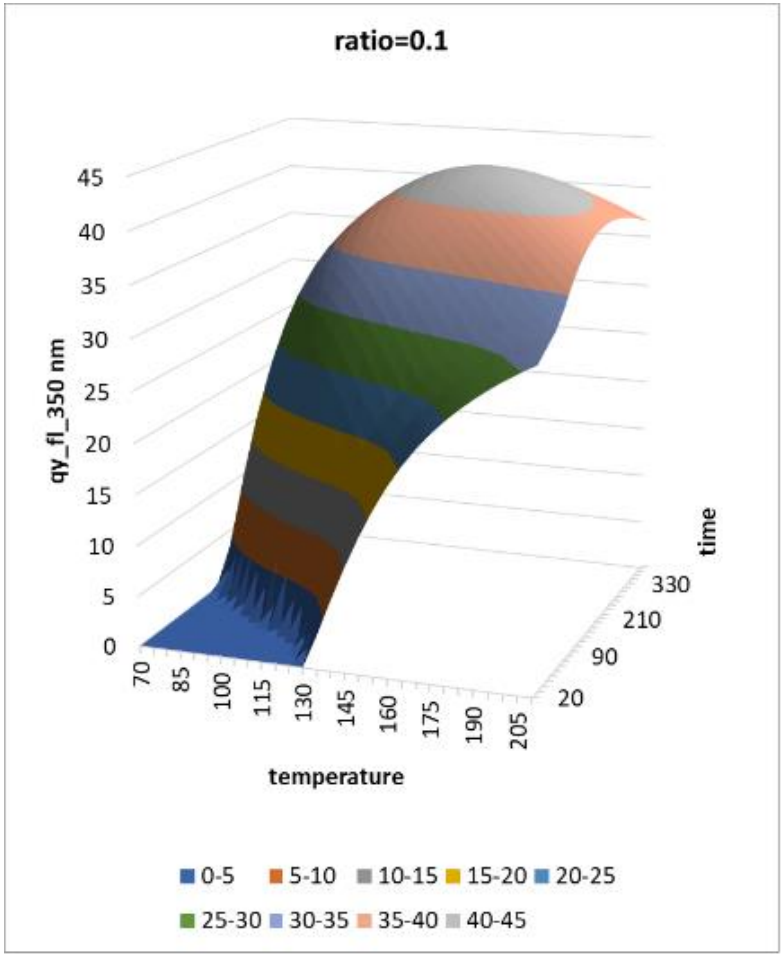
EDA/CA=2:1, time of synthesis increases from left to right, temperature of synthesis from top to bottom.

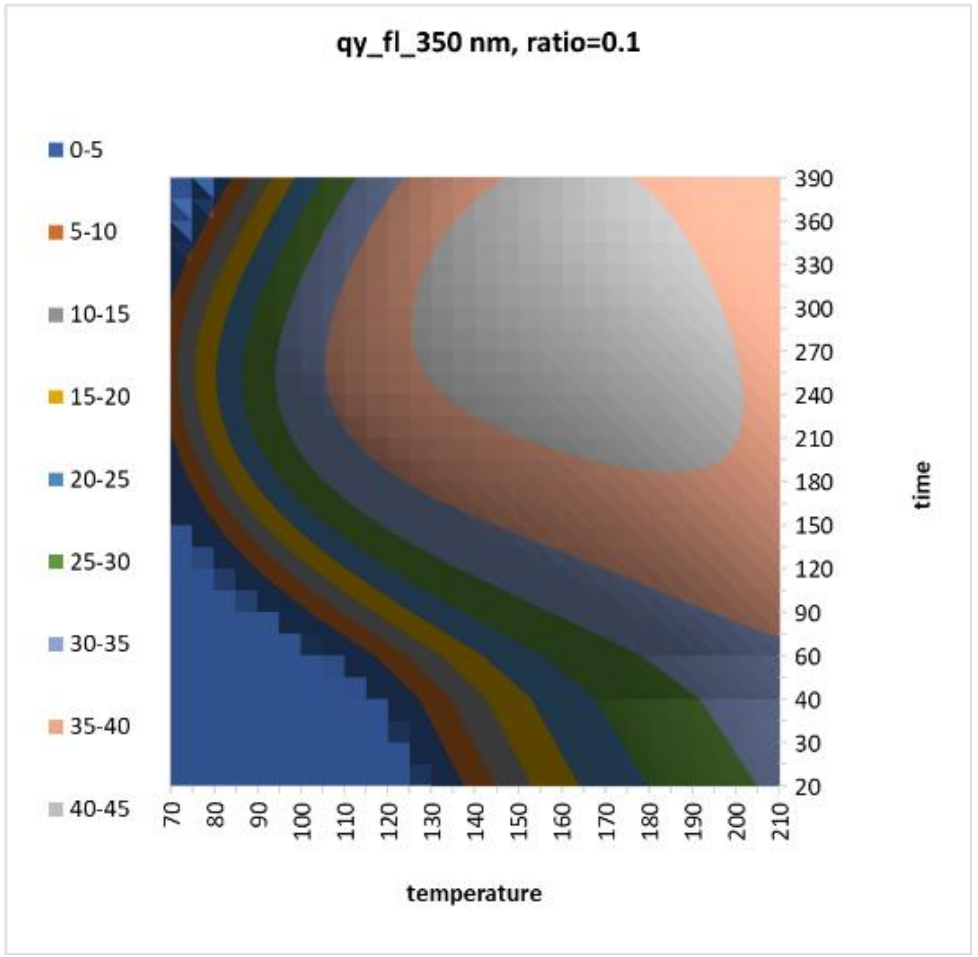
Neural Network Interpolation of the Synthesis Parameters – CD QYL Dependence

Optimal set of neural network parameters:

- Multi-layer perceptron, single hidden layer with 8 neurons
- Number of weights << Number of patterns,
  therefore no overtraining
- Error backpropagation, SGD, batch size=1
- Learning rate 0.01, momentum 0.5
- Logistic activation in hidden layer, linear in the output one
- Stop training after 500 epochs with no training loss reduction

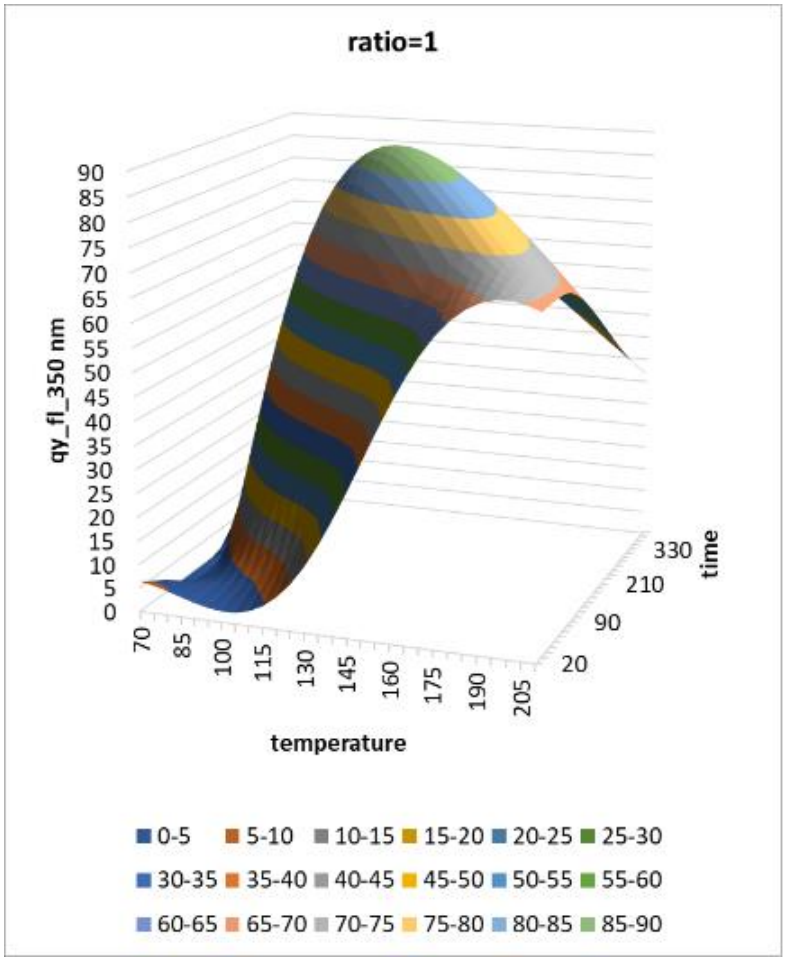
Neural Network Interpolation of the Synthesis Parameters – CD QYL Dependence

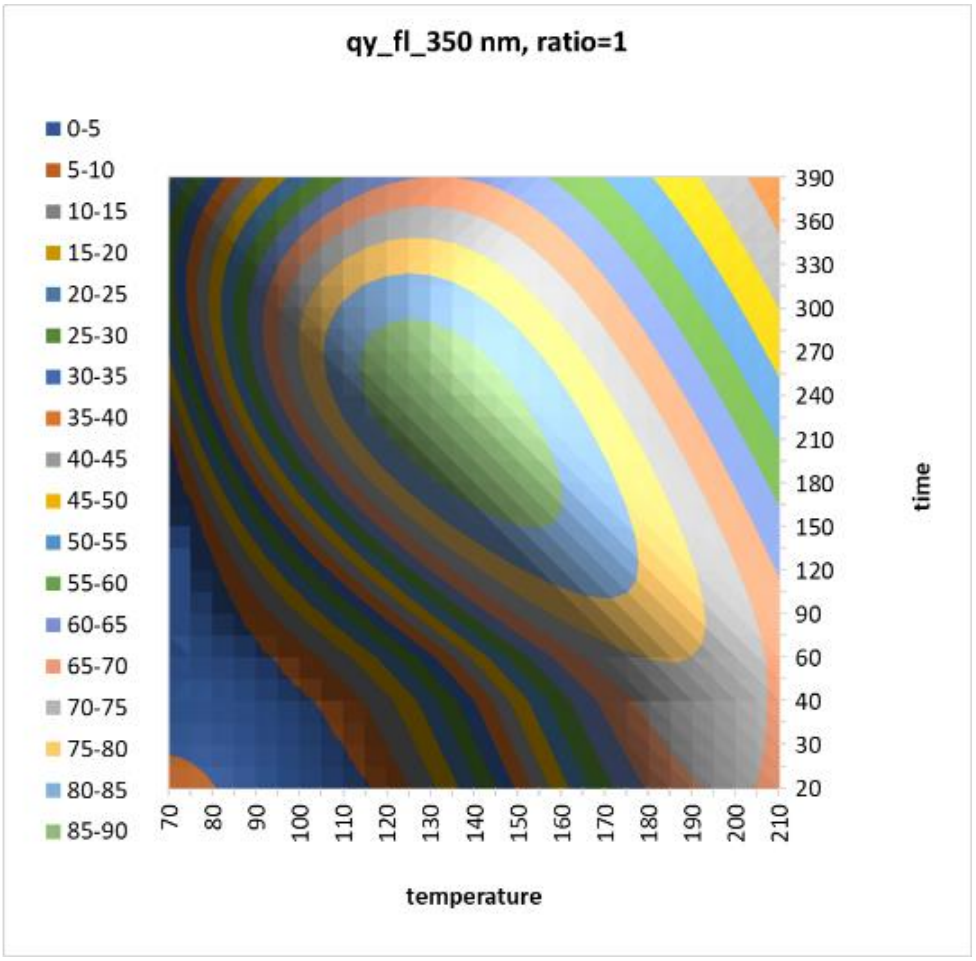




CD QYL vs (Time, Temperature) at EDA/CA=0.1

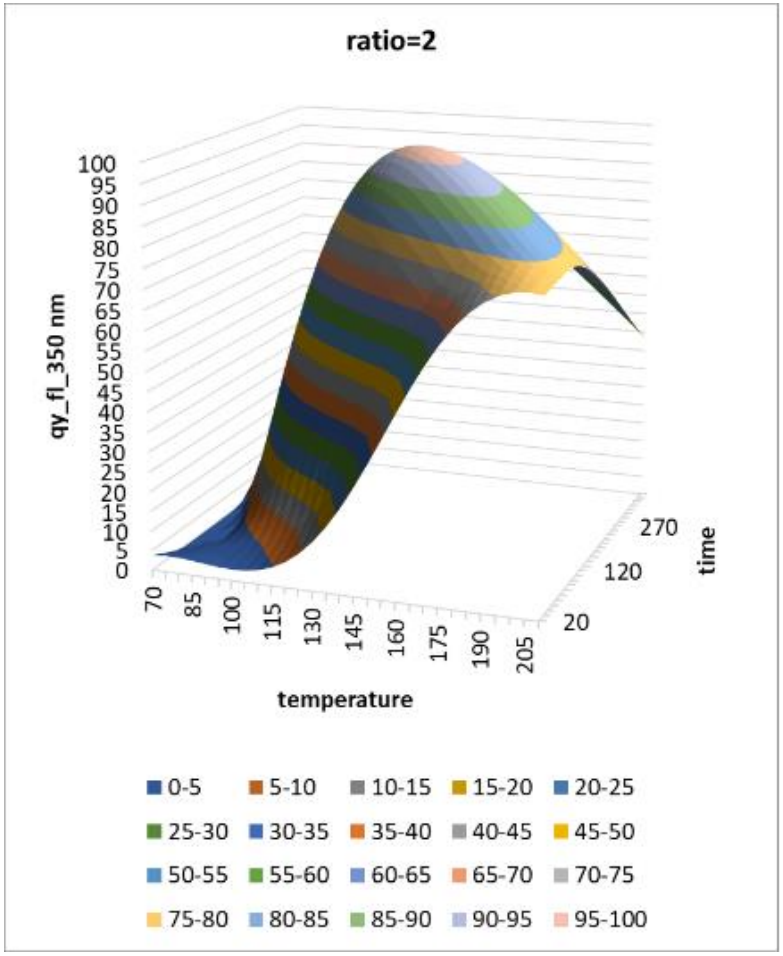
Neural Network Interpolation of the Synthesis Parameters – CD QYL Dependence

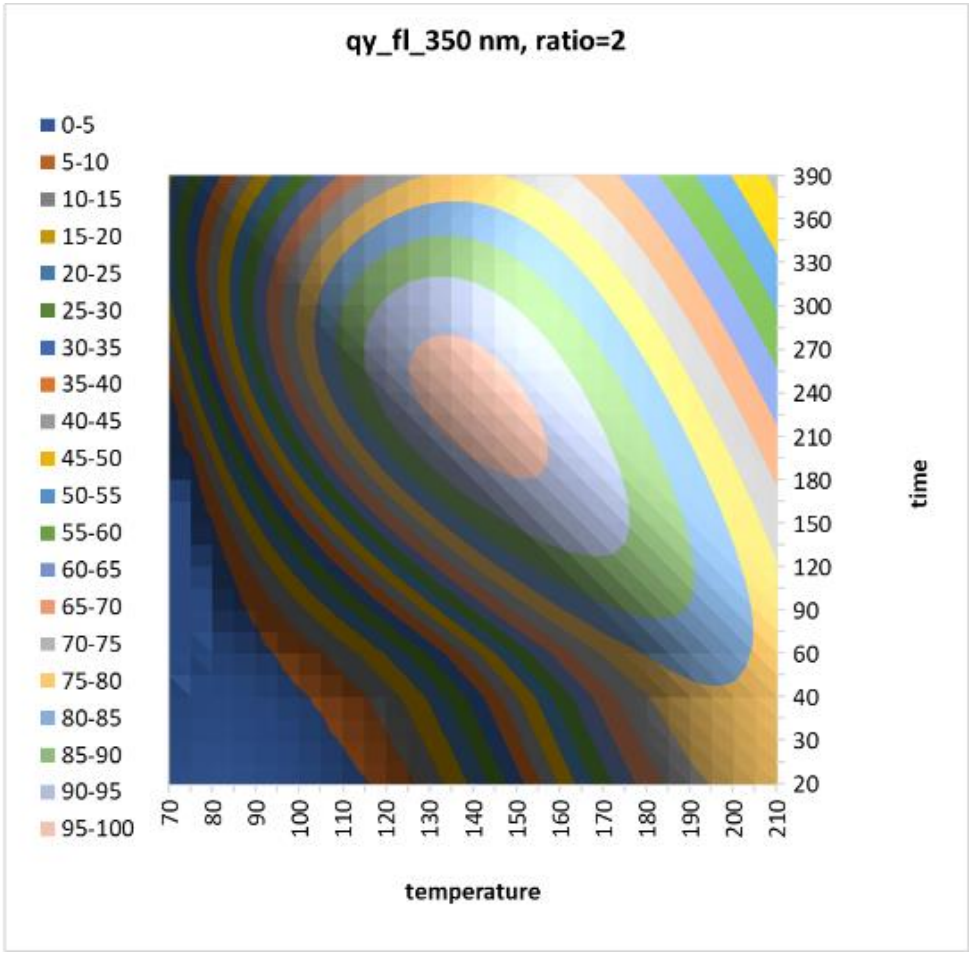




CD QYL vs (Time, Temperature) at EDA/CA=1

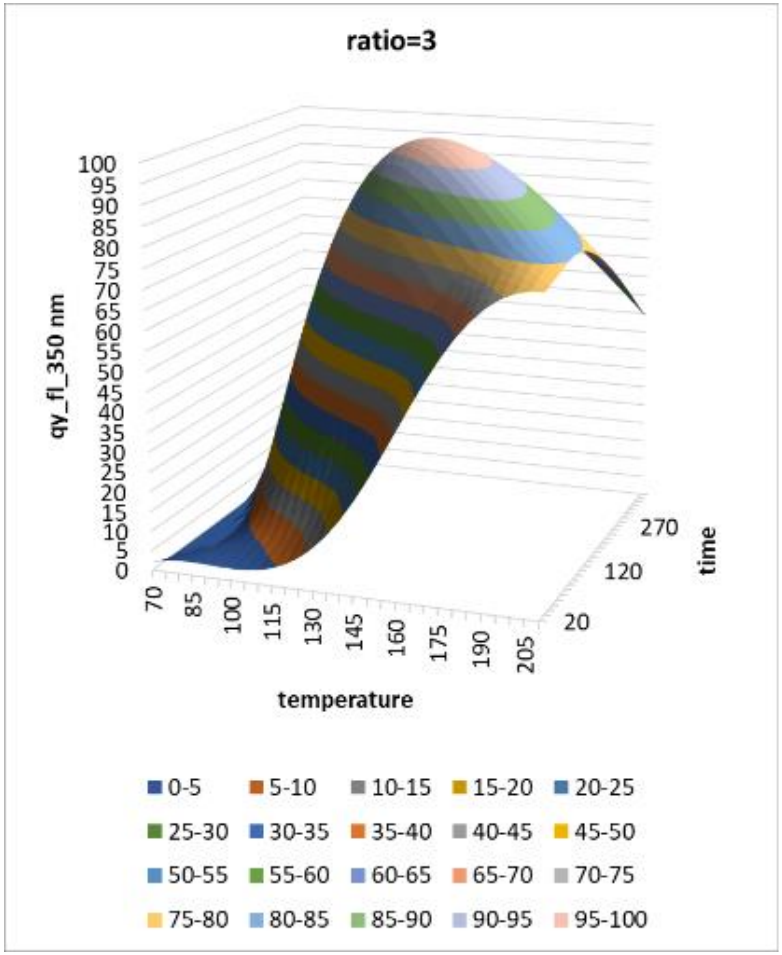
Neural Network Interpolation of the Synthesis Parameters – CD QYL Dependence

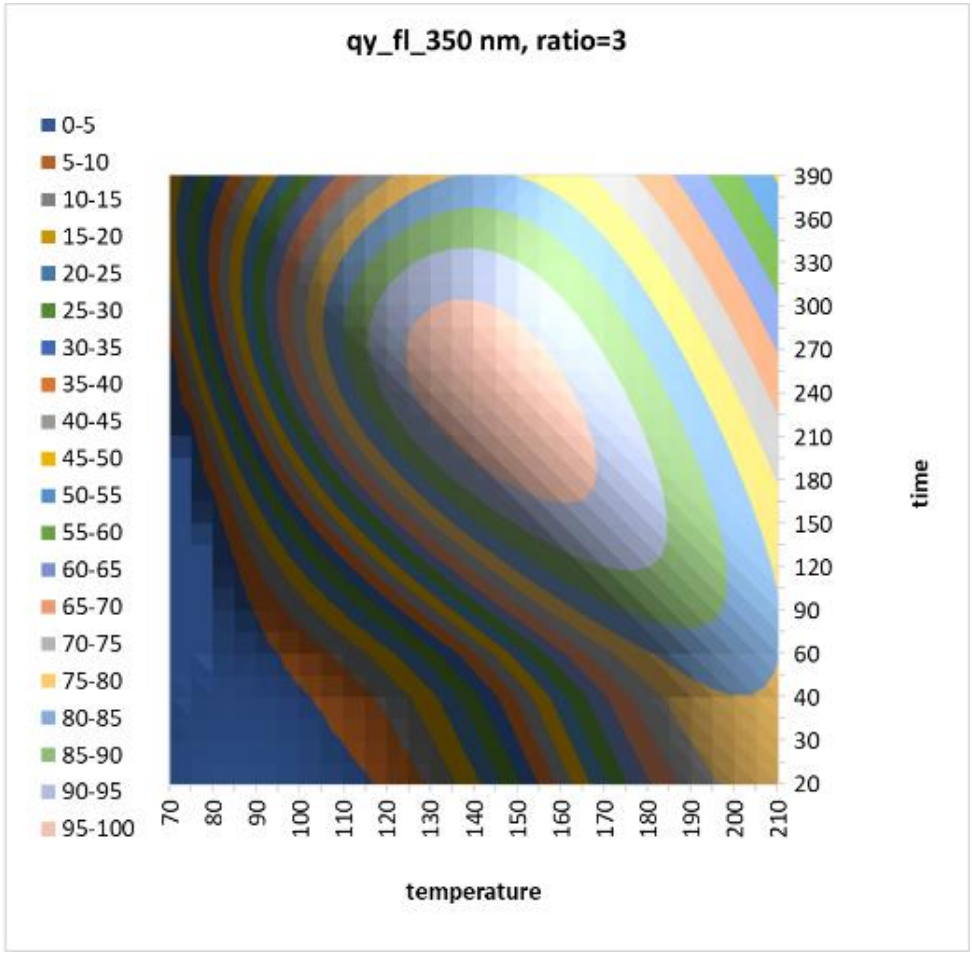




CD QYL vs (Time, Temperature) at EDA/CA=2

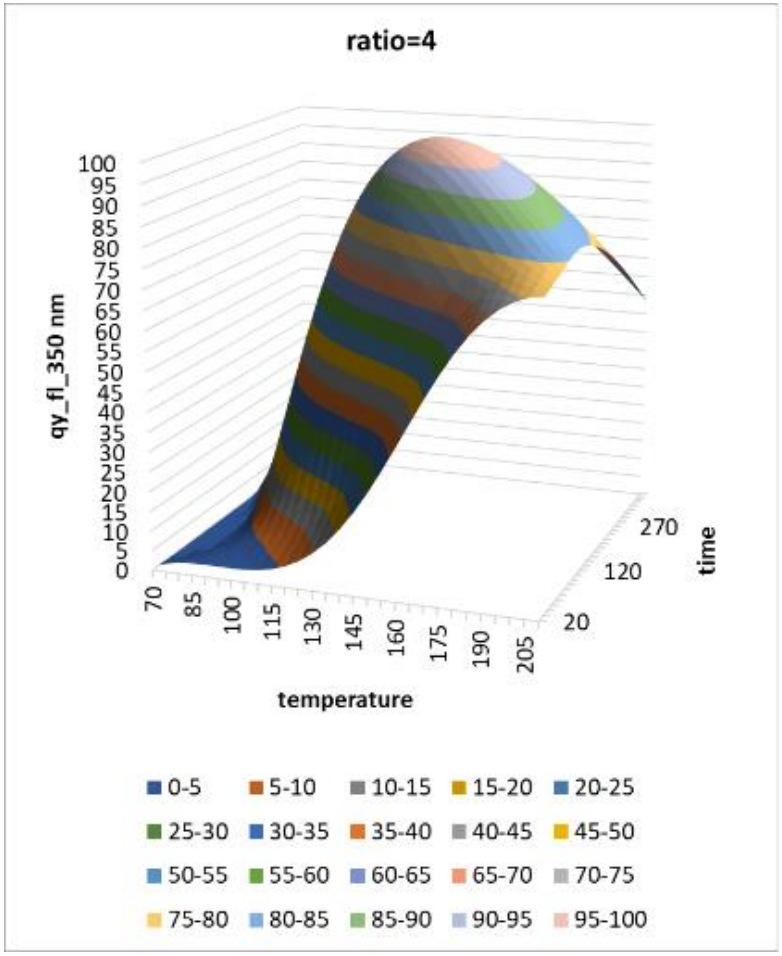
Neural Network Interpolation of the Synthesis Parameters – CD QYL Dependence

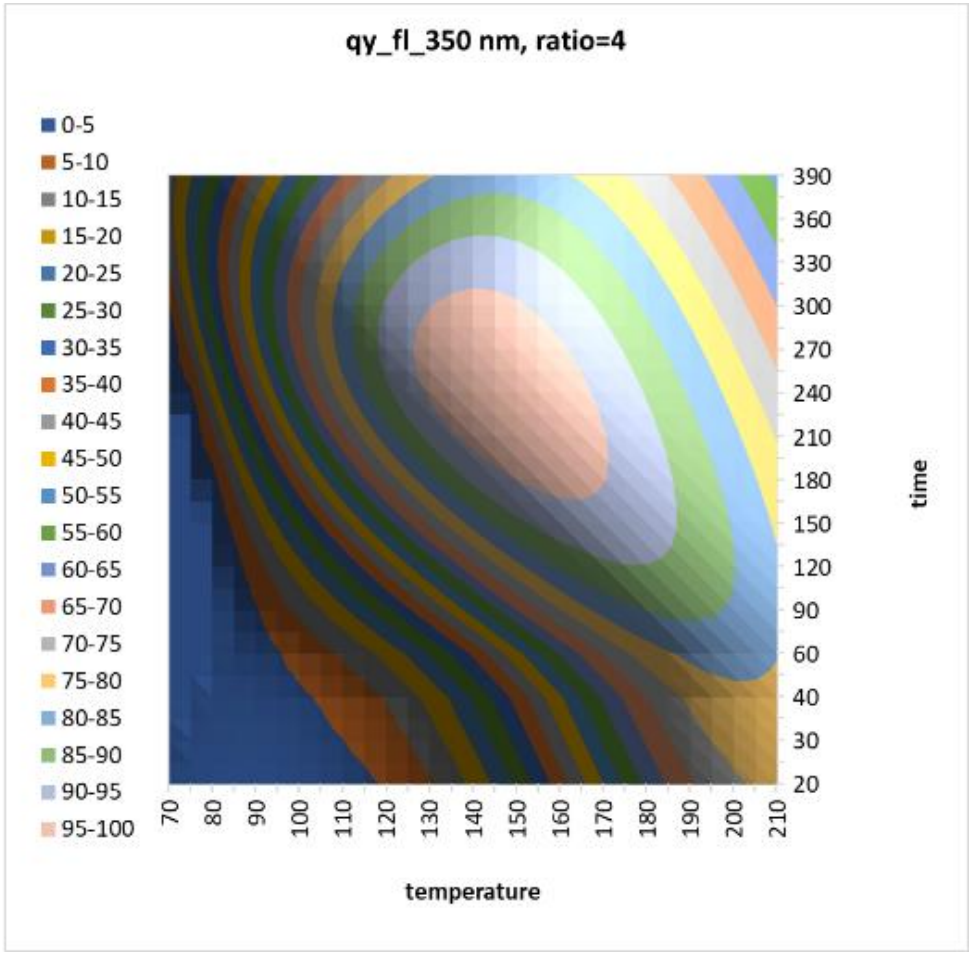




CD QYL vs (Time, Temperature) at EDA/CA=3

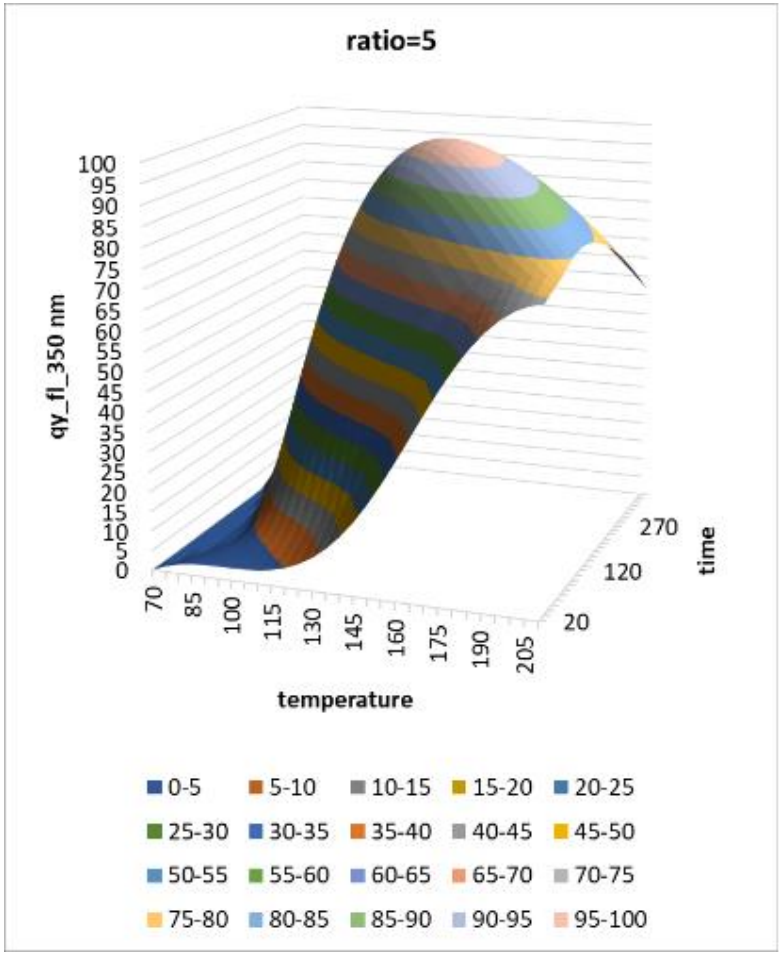
Neural Network Interpolation of the Synthesis Parameters – CD QYL Dependence

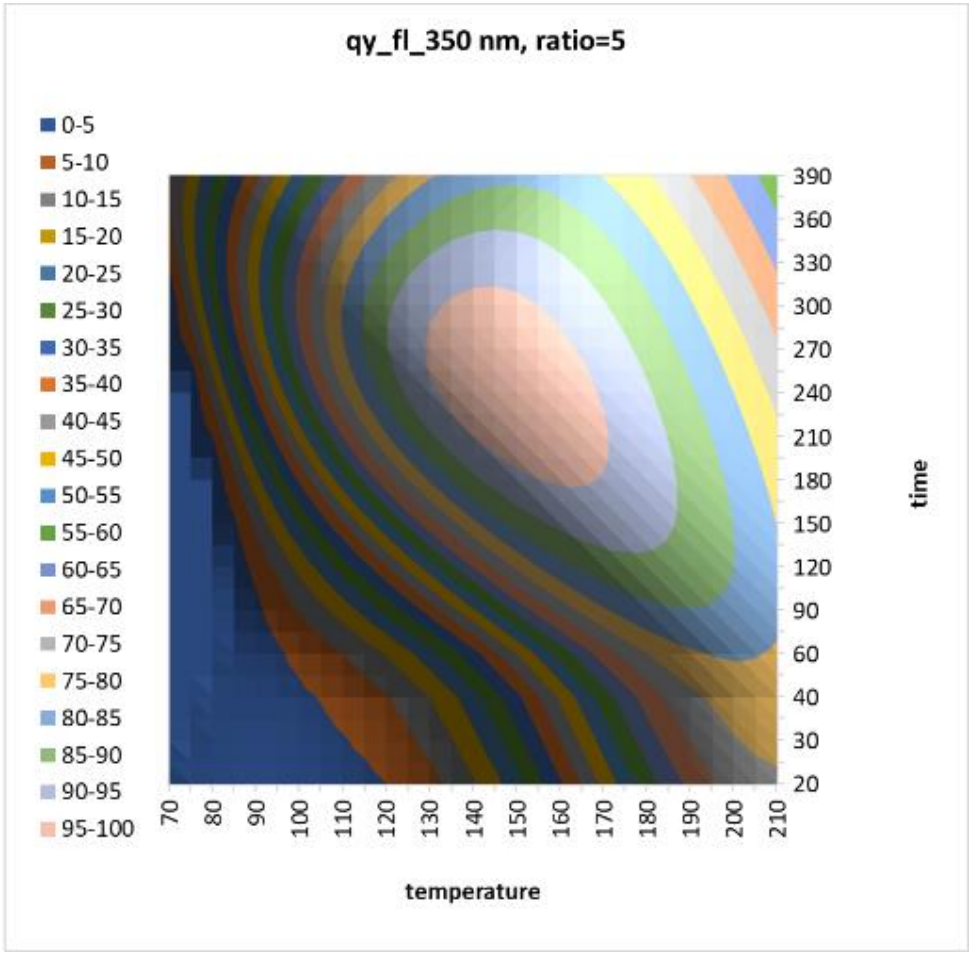




CD QYL vs (Time, Temperature) at EDA/CA=4

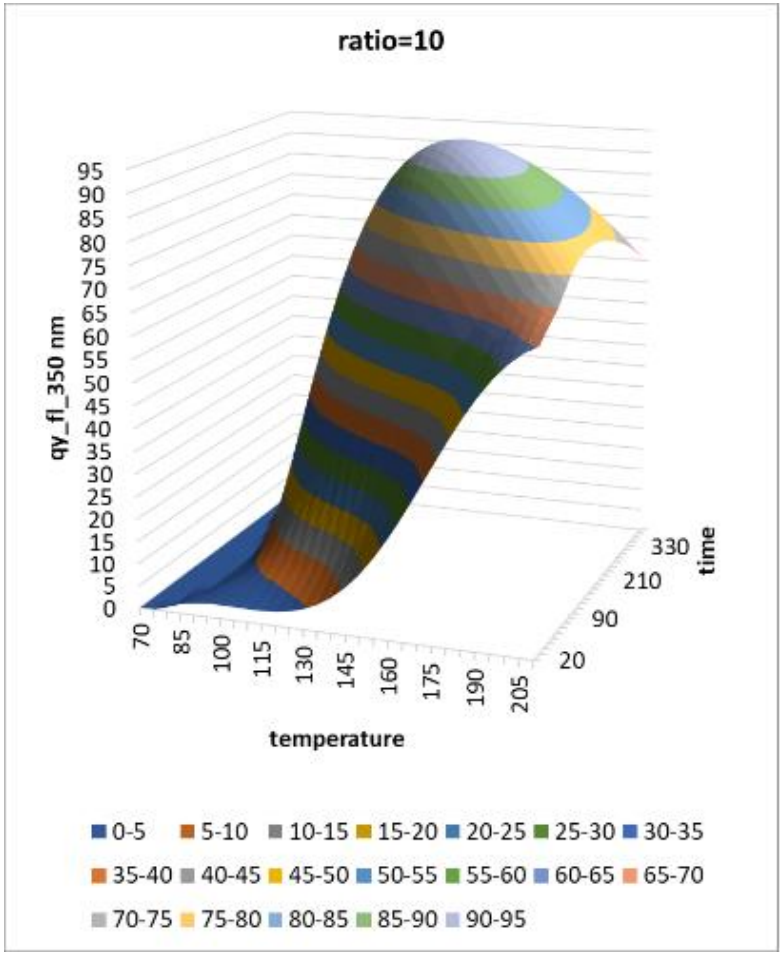
Neural Network Interpolation of the Synthesis Parameters – CD QYL Dependence

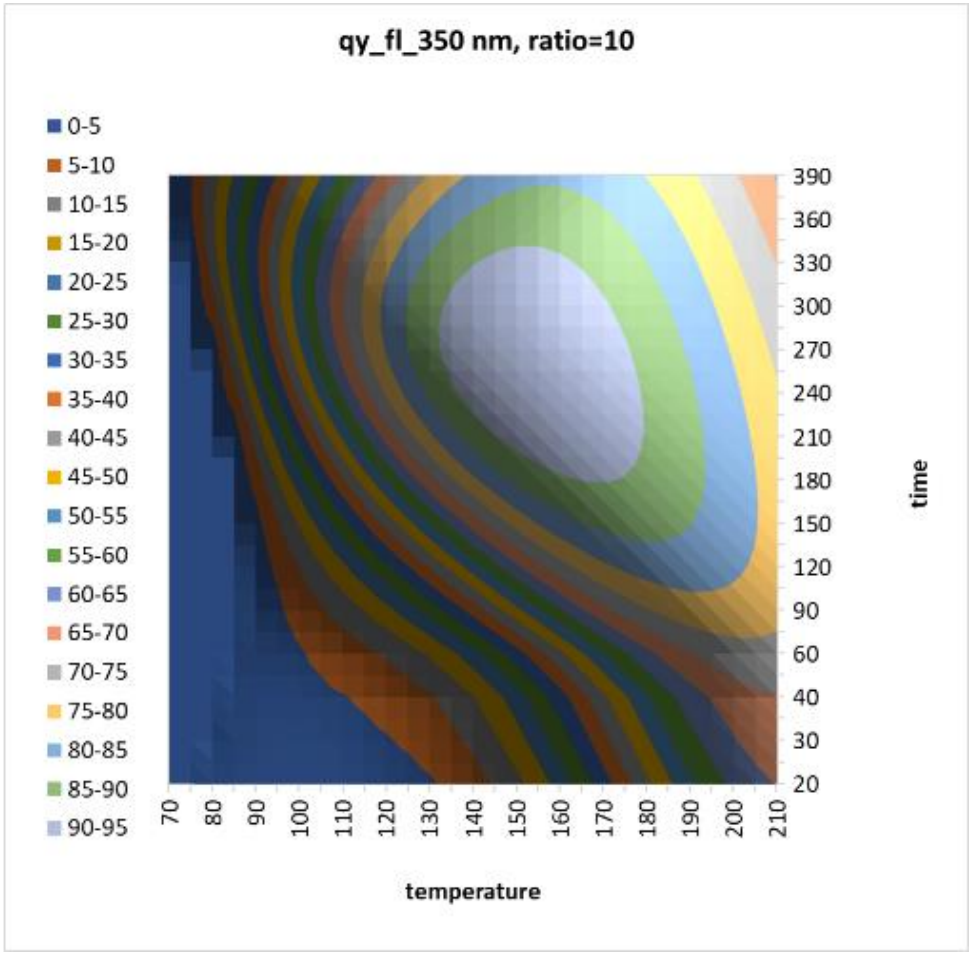




CD QYL vs (Time, Temperature) at EDA/CA=5

Neural Network Interpolation of the Synthesis Parameters – CD QYL Dependence





CD QYL vs (Time, Temperature) at EDA/CA=10

Determination of the Optimal Parameter Set to Obtain the Highest Quantum Yield

Optimal set of synthesis parameters:

- Precursor ratio EDA/CA=3.5
- Synthesis temperature T=145 °C
- Synthesis time t=240 sec

At these parameters, the QYL estimation by the NN was 99.15%

The experimental value of QYL of CD obtained at these parameters was 98.9%

Experimental verification of the NN estimations on a test set:

Maximum QYL error 3.45%, RMSE 2.14%

Conclusions

- The technology of creating carbon dots in laboratory conditions has been successfully implemented, about 355 samples were synthesized.
- To estimate the quantum yield of luminescence of the created CD at given synthesis parameters, a neural network approximator has been created and optimized
- It provided an acceptable approximation error of RMSE=2.14%
- The optimal values of synthesis parameters corresponding to the highest QYL of the synthesized CD were determined
- The highest possible QYL has been estimated as 99.15%, while its experimental value obtained at these parameter values was 98.9%
- A multi-layer perceptron was proved to be an efficient approximator for solution of the QSPR inverse problem.

Thank you for your attention