

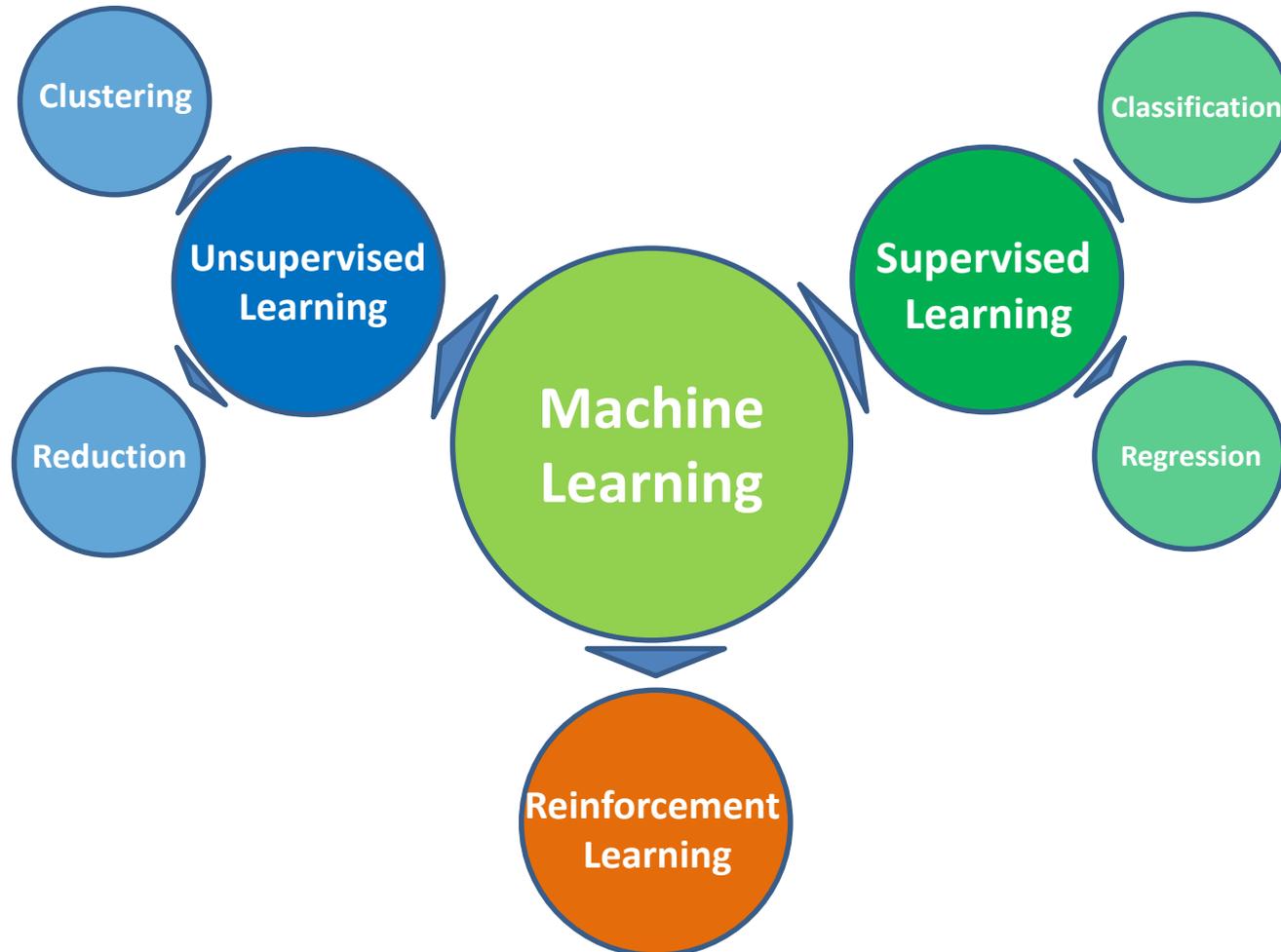


Machine Learning Assisted Discovery of Photoluminescent Materials

Romain GAUTIER

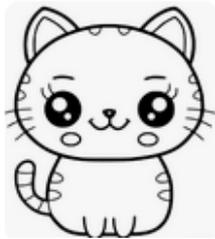
Institut des Matériaux de Nantes Jean Rouxel (IMN)

What is machine learning ?



What is machine learning ?

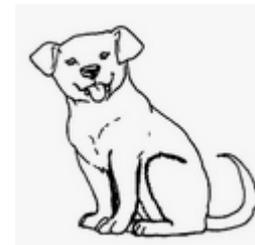
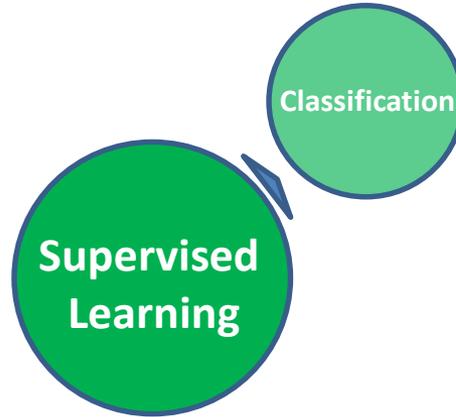
Predict qualitative information



These are cats



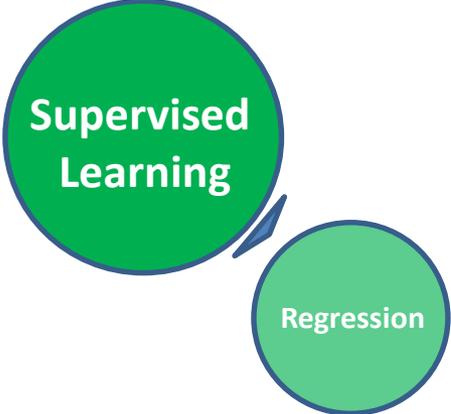
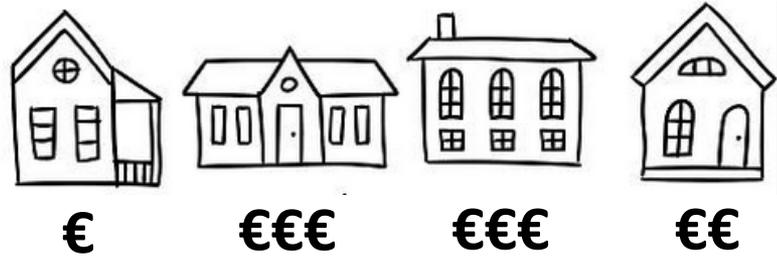
These are dogs



What is this?

What is machine learning ?

Predict quantitative information



What is the price?

I) Classification:

Prediction of the photoluminescence color for $\text{Li}_2\text{BaSiO}_4:\text{Eu,Ce}$

II) Regression:

Prediction of the CIE coordinates of the photoluminescence for $\text{Ca}_{14}\text{Zn}_6\text{Ga}_{10}\text{O}_{35}:\text{Mn,Tm}$

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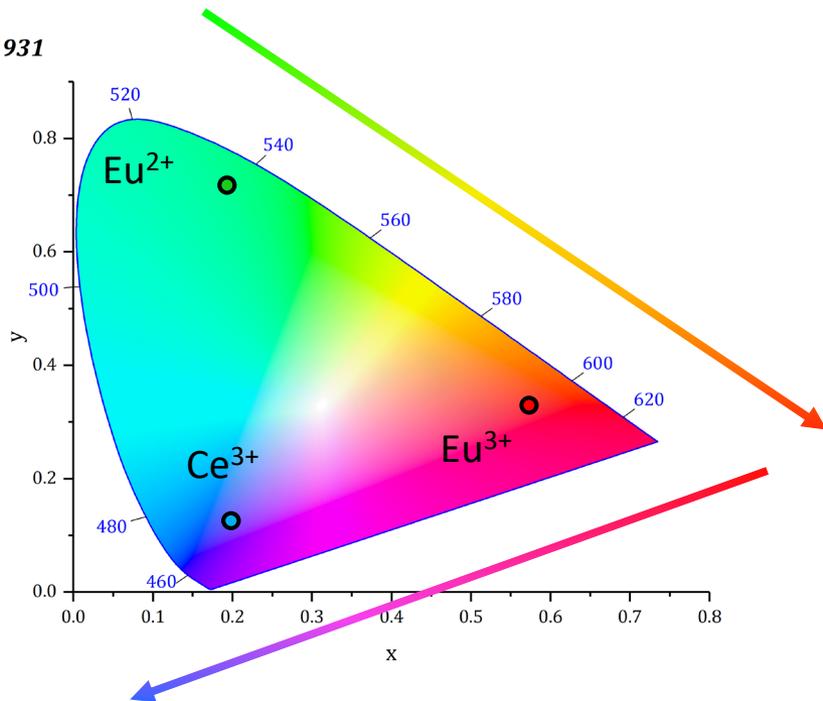
Prediction of the photoluminescence color

Oxide Matrix + Control of luminescent center valence state

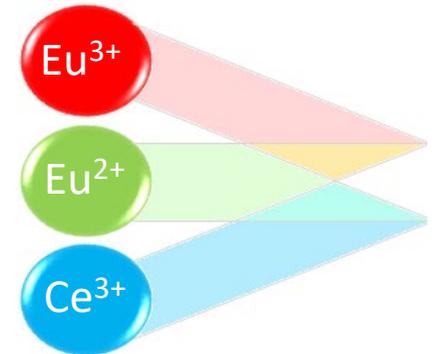


LBSO

CIE 1931



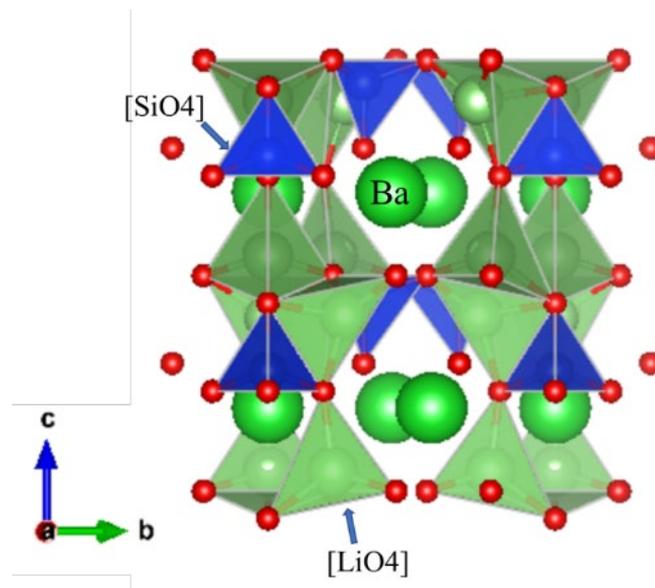
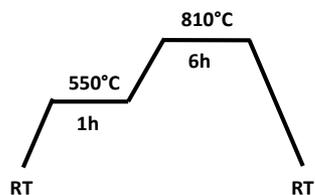
UV excitation



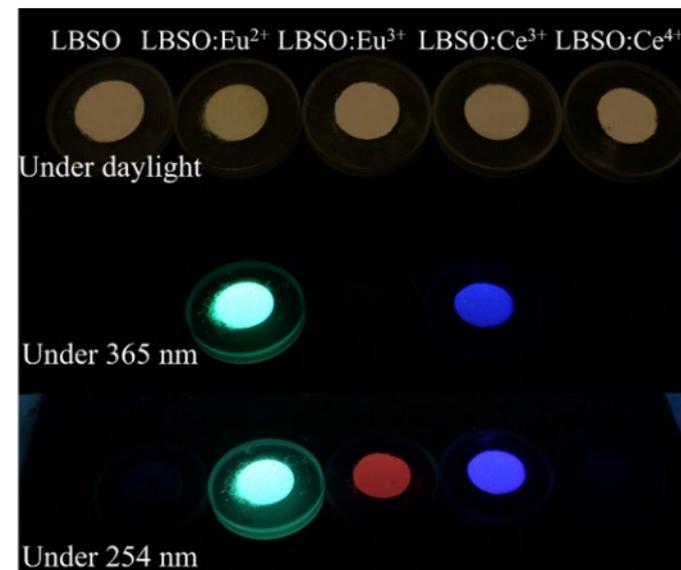
Prediction of the photoluminescence color



Solid-state synthesis



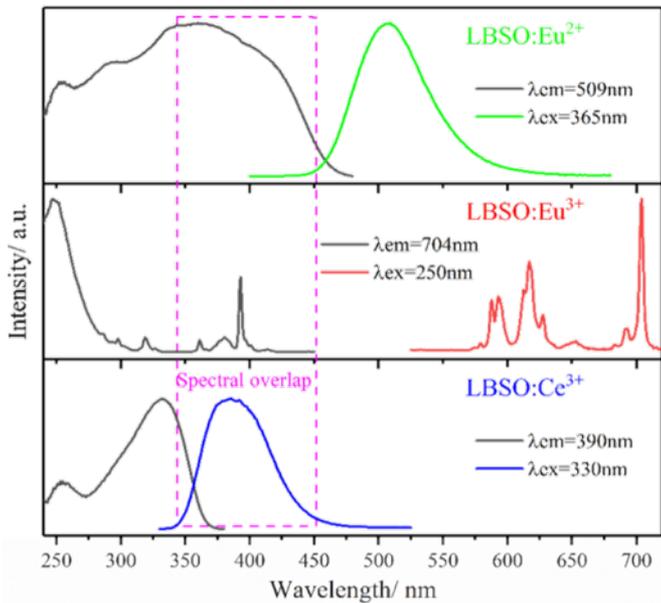
Crystal structure of the LBSO
Space group: $P6_3cm$



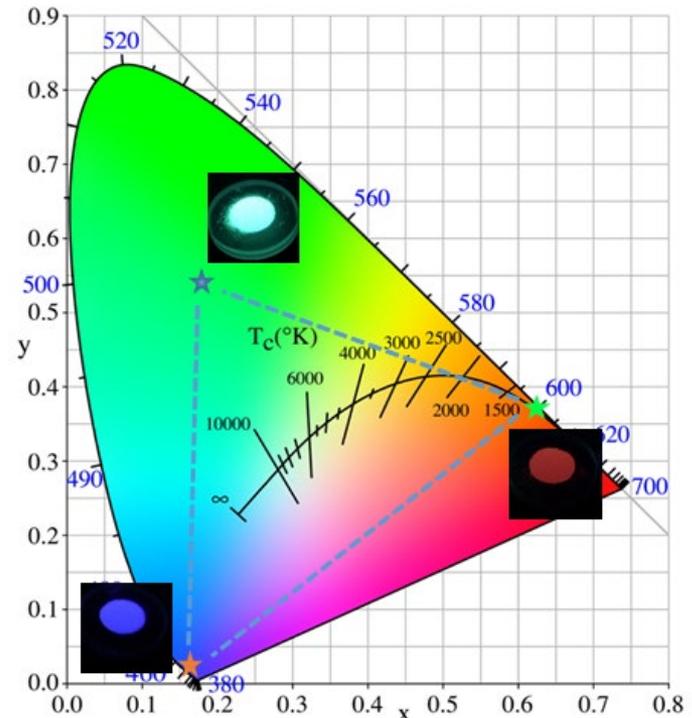
Pictures of the LBSO, LBSO:Eu²⁺, LBSO:Eu³⁺, LBSO:Ce³⁺, LBSO:Ce⁴⁺ samples under daylight and UV lamp ($\lambda = 365 \text{ nm} / 254 \text{ nm}$).

Prediction of the photoluminescence color

$\text{Li}_2\text{BaSiO}_4:\text{Eu,Ce}$
(LBSO:Eu,Ce)



PL and PLE spectra of LBSO:Eu²⁺, LBSO:Eu³⁺ and LBSO:Ce³⁺ samples.



CIE coordinates of LBSO:Eu²⁺, LBSO:Eu³⁺ and LBSO:Ce³⁺

Prediction of the photoluminescence color

Predict qualitative information



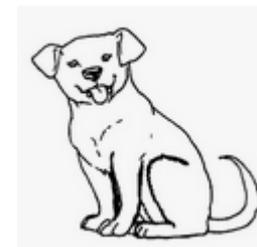
These are cats



These are dogs

Supervised
Learning

Classification



What is this?

Prediction of the photoluminescence color

Predict qualitative information

Material 1 Material 2 Material 3
Conditions *Conditions* *Conditions*

These are blue phosphors

Material 4 Material 5 Material 6
Conditions *Conditions* *Conditions*

These are yellow phosphors

Supervised
Learning

Classification

Material 7

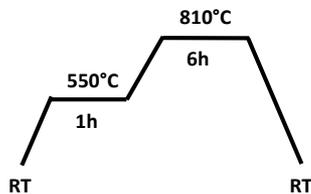
Conditions

What is this?

Prediction of the photoluminescence color

Synthesis

**Solid-state synthesis
under reducing
atmosphere
(95%Ar, 5%H₂)**



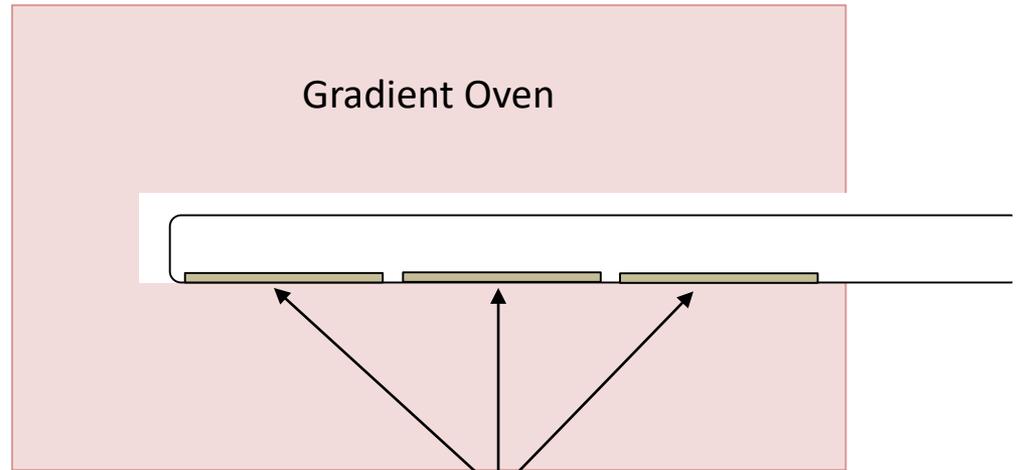
Oxidation thermal treatment

High temperature ←

→ Low Temperature



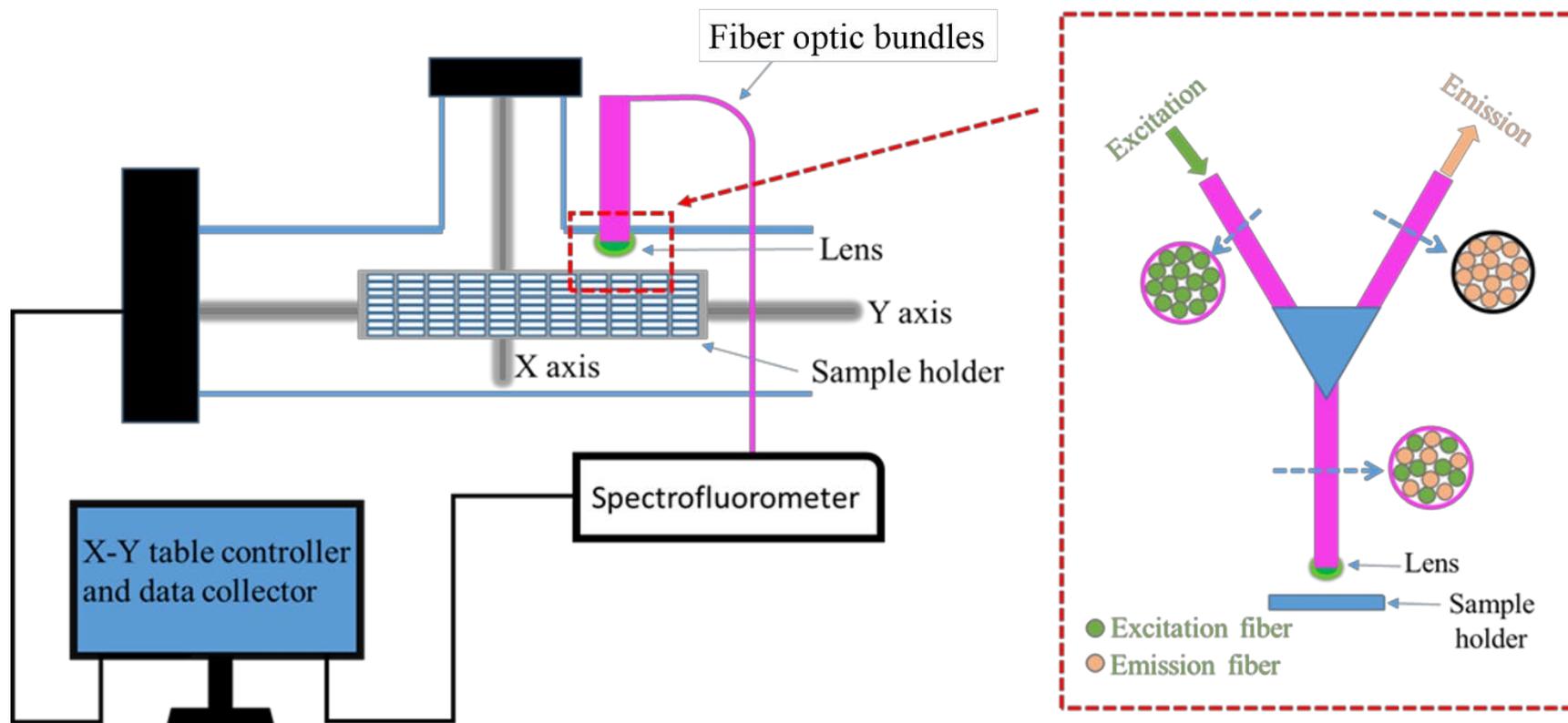
Gradient Oven



Sample holder

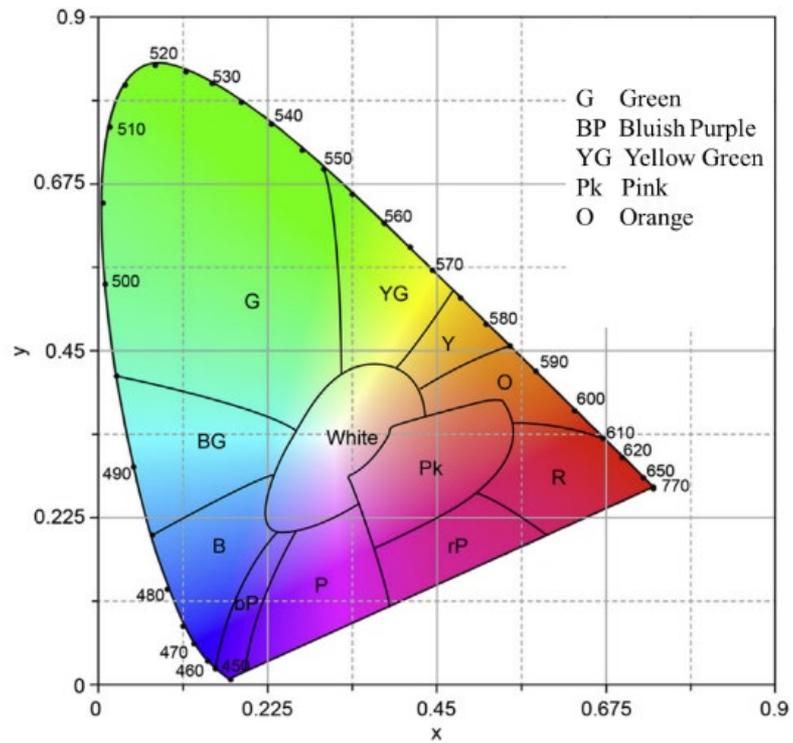
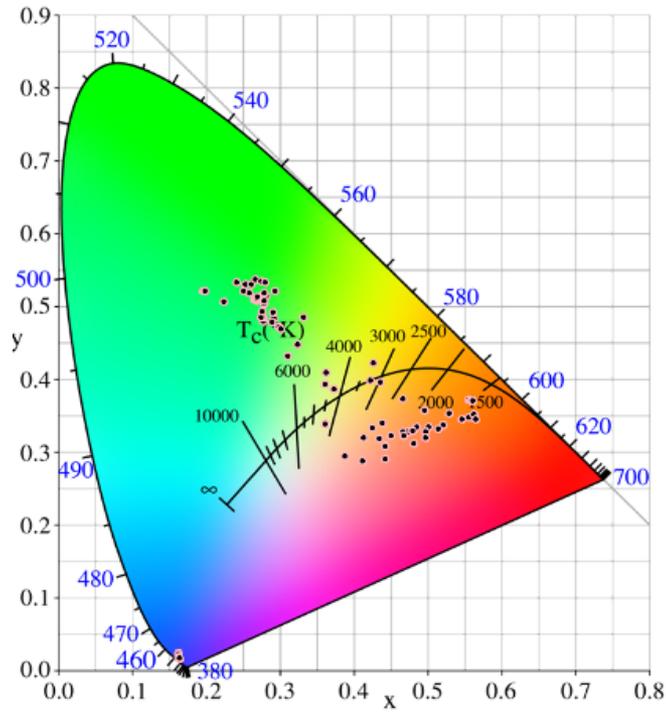
Prediction of the photoluminescence color

Characterization



Prediction of the photoluminescence color

Machine learning to visualize the experimental conditions to target the white luminescence



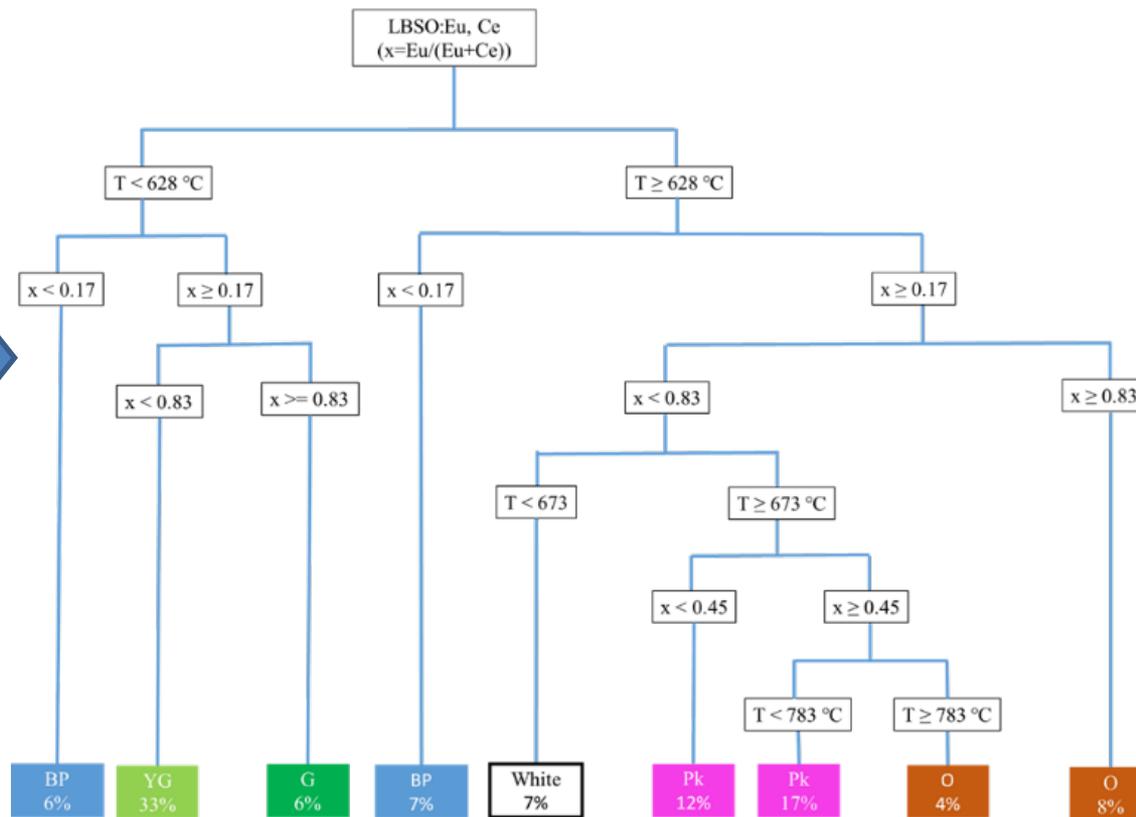
CIE coordinates of the 88 samples

Color names of light sources from KS A 0012 (2013) standard



Prediction of the photoluminescence color

Machine learning to visualize the experimental conditions to target the white luminescence

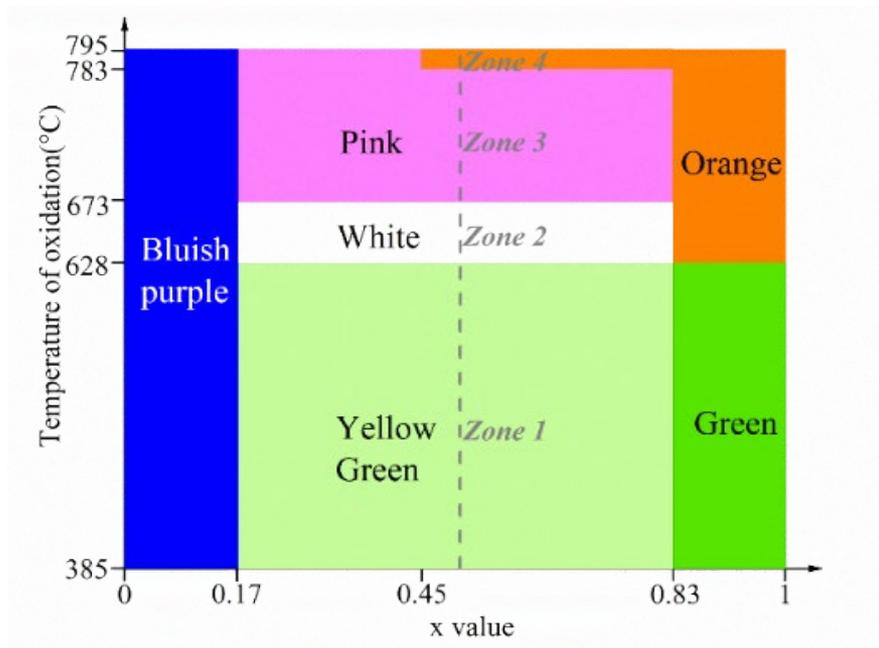


Decision tree

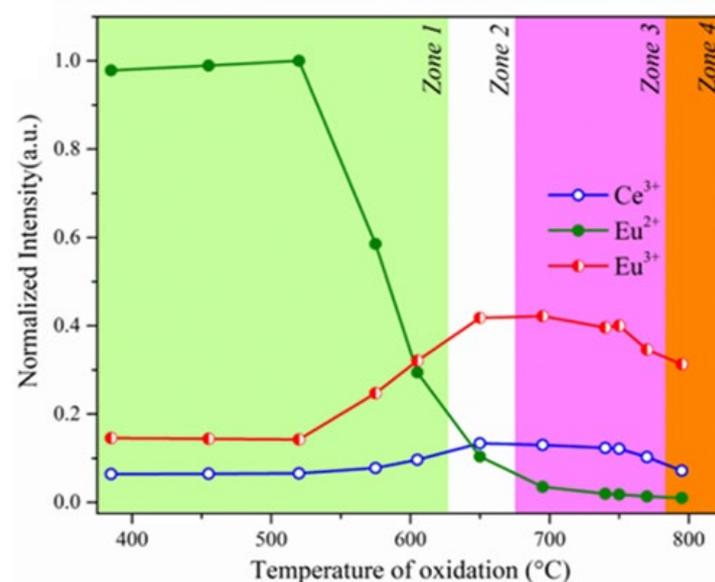
- Dataset
80% for training
20% for test

Prediction of the photoluminescence color

Machine learning to identify the experimental conditions and rationalize the photoluminescence colors



Results of decision tree model based on the 88 samples



Evolution of the emission intensities associated with Ce³⁺ ($\lambda_{em} = 400$ nm, blue line), Eu²⁺ ($\lambda_{em} = 510$ nm, green line), and Eu³⁺ ($\lambda_{em} = 704$ nm, red line) vs. the temperature of oxidation for LBSO:0.5%Eu, 0.5%Ce

I) Classification:

Prediction of the photoluminescence color for $\text{Li}_2\text{BaSiO}_4:\text{Eu,Ce}$

II) Regression:

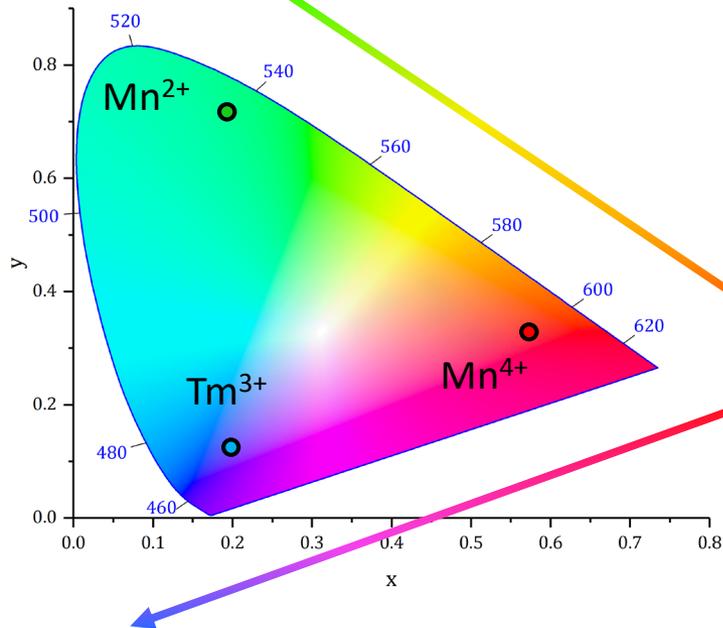
Prediction of the CIE coordinates of the photoluminescence for $\text{Ca}_{14}\text{Zn}_6\text{Ga}_{10}\text{O}_{35}:\text{Mn,Tm}$

Prediction of the CIE coordinates

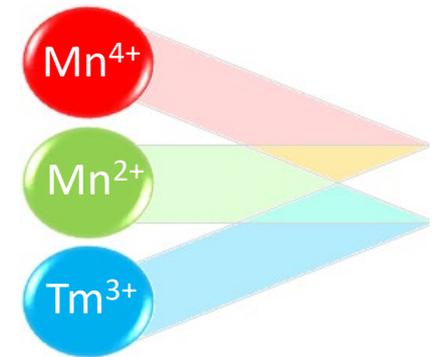


CZGO

CIE 1931



UV excitation



Prediction of the CIE coordinates



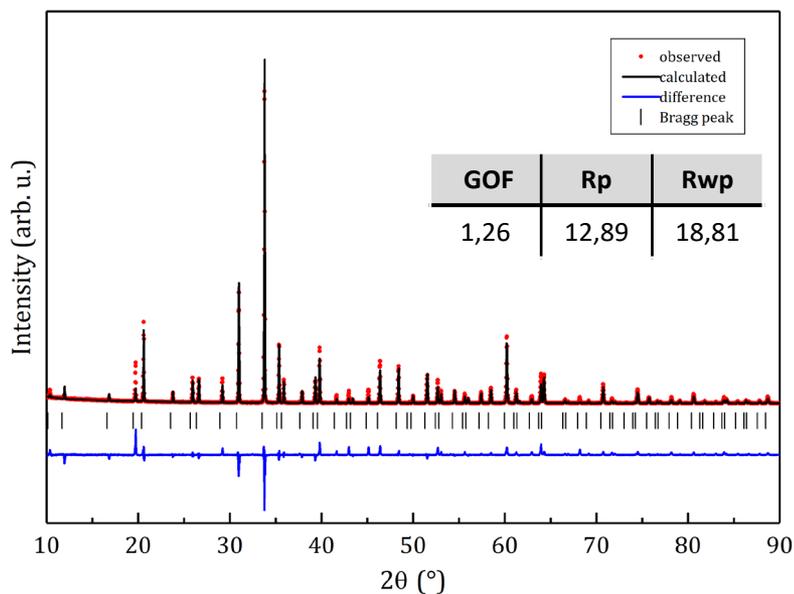
Solid State Synthesis

1250°C 6h

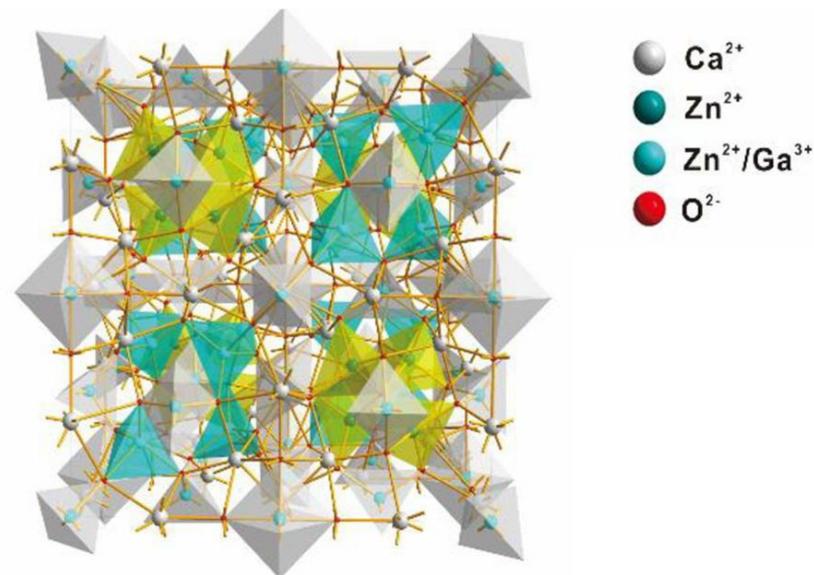


Reductive Thermal Treatment

H₂/Ar



ICSD-245650



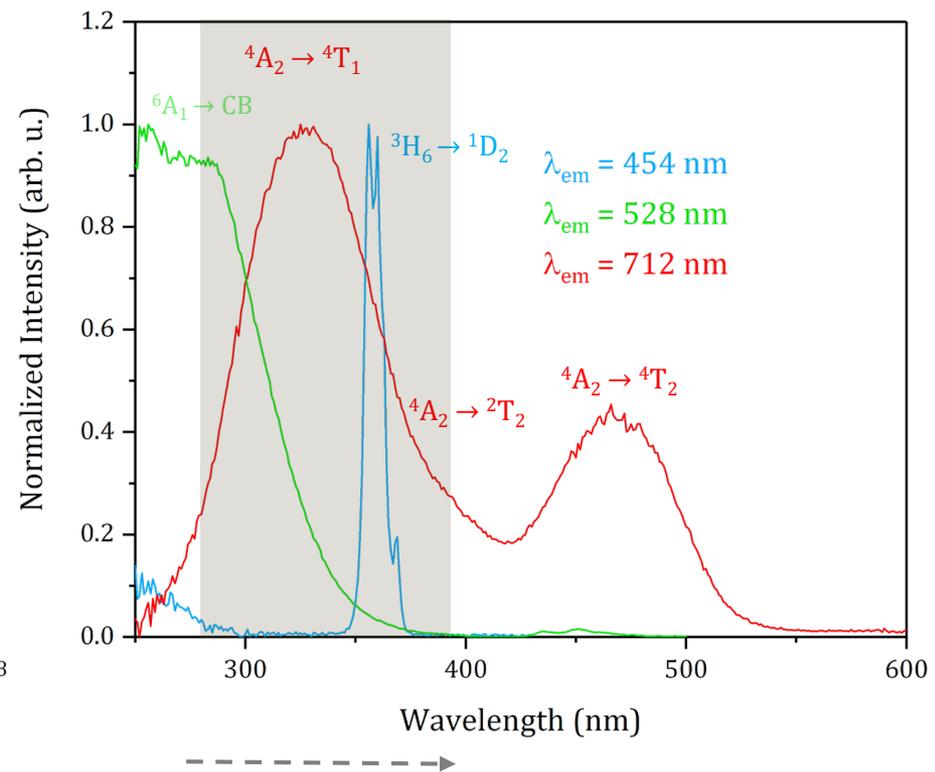
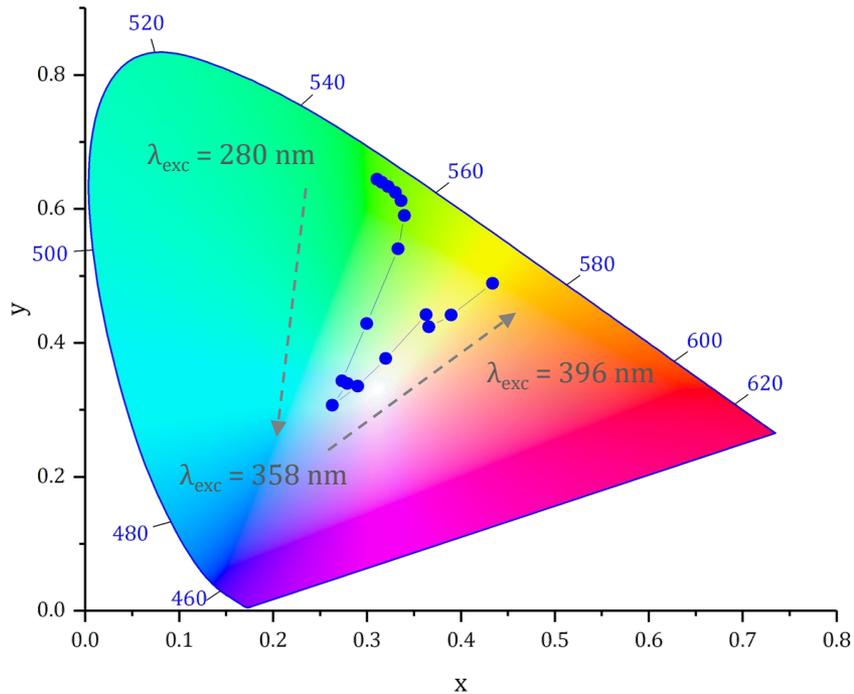
Liao *et al.*, Chemical Engineering Journal 395 (2020) 125060

Cubic Structure
F 4 3 2 (209) space group
a = 15.0556(1) Å

Prediction of the CIE coordinates

CIE coordinates changes according to the excitation wavelength

CIE 1931



What is machine learning ?

Predict quantitative information



Supervised Learning

Regression



What is the price?

What is machine learning ?

Predict quantitative information

Material 1	Material 2	Material 3
<i>Conditions</i>	<i>Conditions</i>	<i>Conditions</i>
(x_1, y_1)	(x_2, y_2)	(x_3, y_3)
Material 4	Material 5	Material 6
<i>Conditions</i>	<i>Conditions</i>	<i>Conditions</i>
(x_4, y_4)	(x_5, y_5)	(x_6, y_6)

Supervised Learning

Regression

Material 7

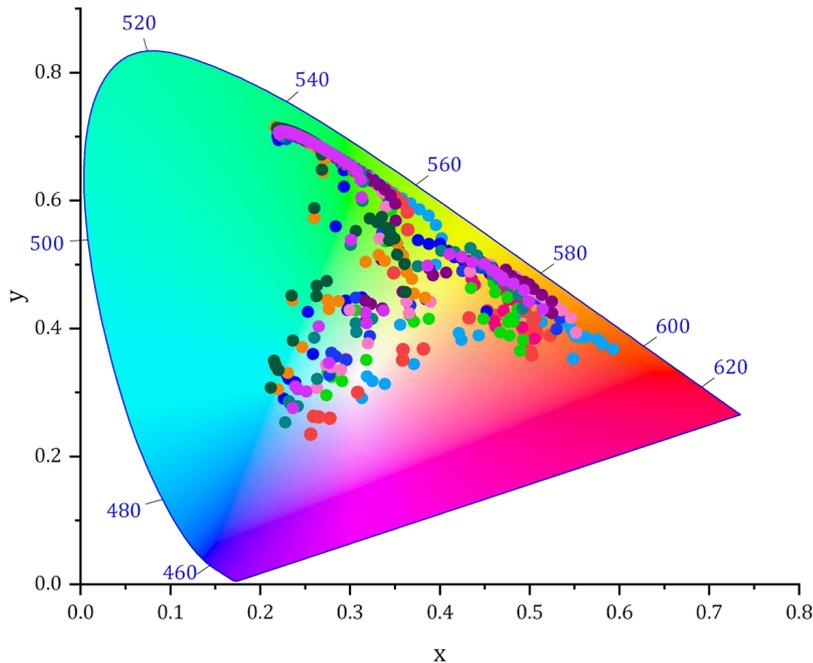
Conditions

What is the (x, y) ?

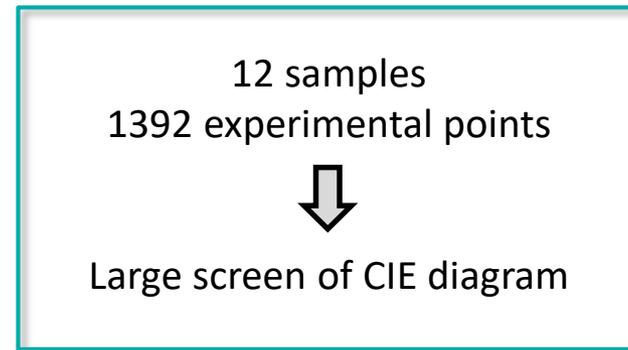
Prediction of the CIE coordinates

Parameters / sample n°	1	2	3	4	5	6	7	8	9	10	11	12
[Mn]	1	1,1	1,1	1,5	1,5	1	1	1,5	1,5	1,5	1	1
[Tm]	2	2,8	2,8	3	3	1,5	1,5	1,5	6	6	3	3
Duration of TTR at 900°C (h)	1	1	12	12	6	2	12	12	12	24	2	6
λ_{exc} (nm)	280-396	280-396	280-396	280-296	280-296	280-296	280-296	280-296	280-296	280-296	280-296	280-296

CIE 1931



4 parameters for the modulation of the resulting emission color

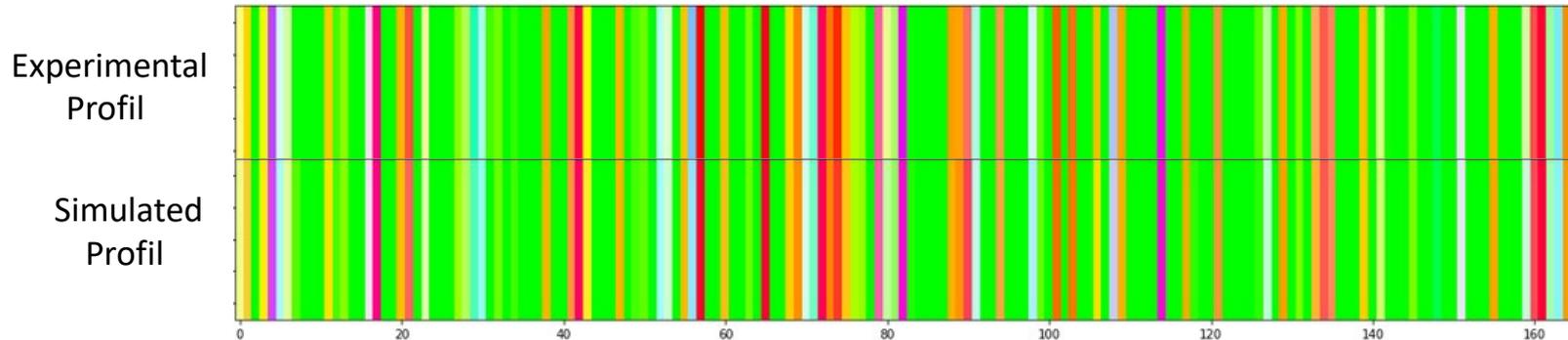


➔ **XGBoost model (eXtreme Gradient Boosting)**

Prediction of the CIE coordinates

- ✓ Validation on 20% of the resulting data (= 165 experimental points)

Representation of the resulting emission color



- ✓ Experimental validation

Input parameters

[Mn] : 1 - 1.5 % - step 0.1 %

[Tm] : 1 - 6 % - step 0.1 %

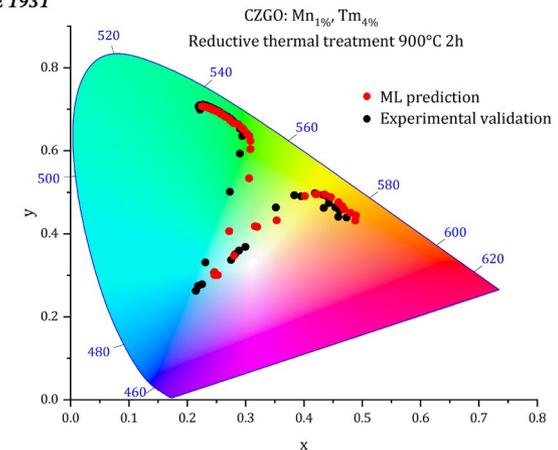
Temperature of RTT : 900 °C

Time of RTT : 1 - 24 h - step 1 h

λ_{exc} : 280 - 396 nm - step 2 nm

> 7000 combinations

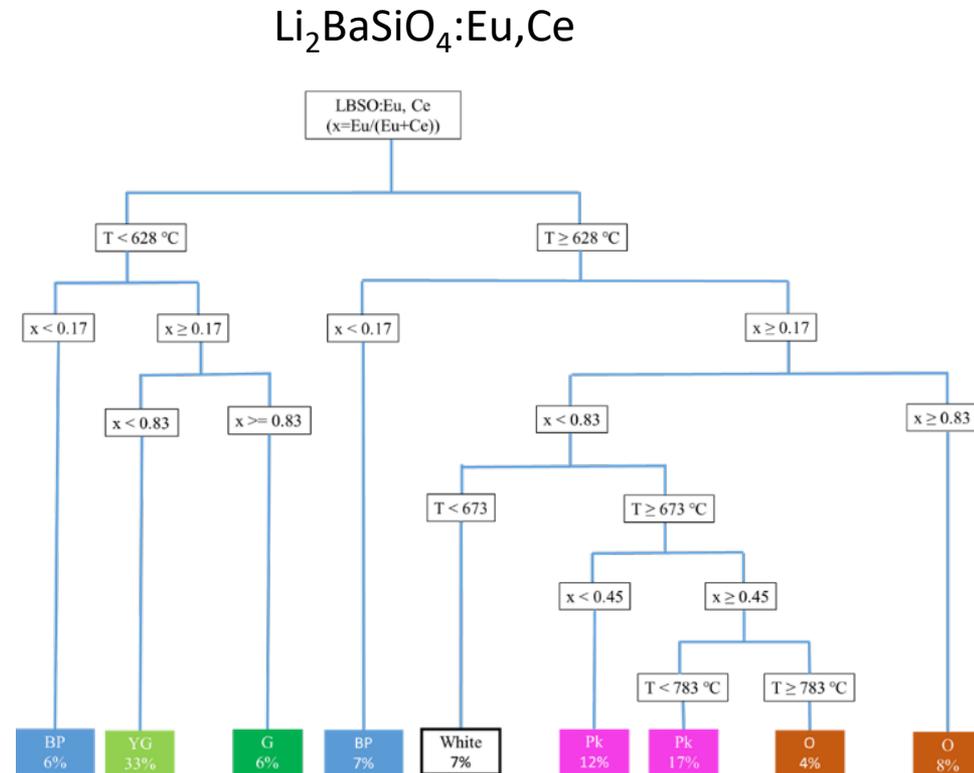
CIE 1931



Conclusion

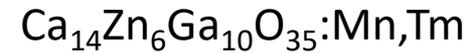
Take-home messages

- ML to identify the key parameters influencing the property.

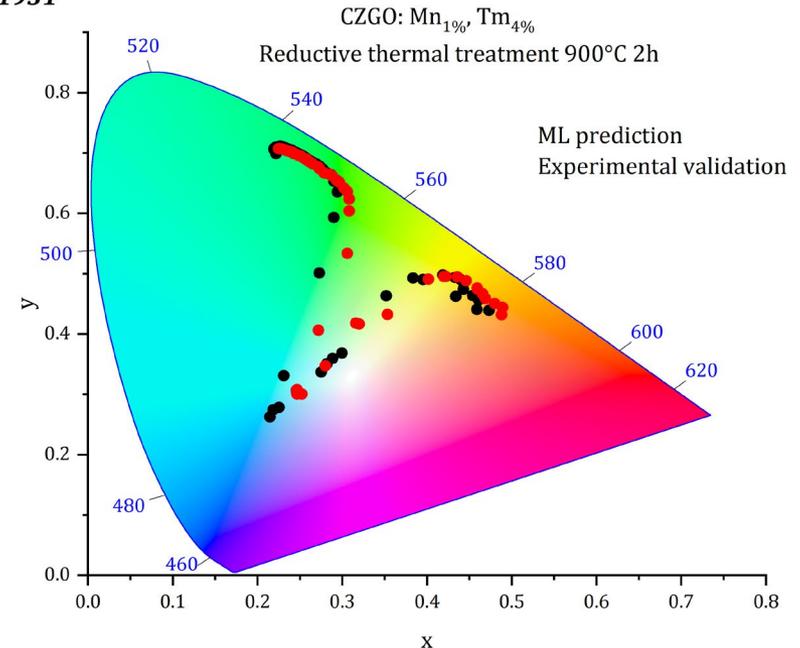


Take-home messages

- ML to identify the key parameters influencing the property.
- ML to predict accurately the property as function of multiple variables.



CIE 1931



Acknowledgment

Romain Génois
PhD student

Hailong Yuan
PhD student

Estelle Glais
Postdoc

Luyuan Qi
Research scientist

Florian Massuyeau
CNRS Engineer

Fundings:



THANK YOU FOR YOUR ATTENTION

An endless search with short term challenges

The periodic table of elements

1 IA																	18 VIIIA				
1 H Hydrogen 1.008																	2 He Helium 4.002602				
3 Li Lithium 6.94	4 Be Beryllium 9.012182															5 B Boron 10.81	6 C Carbon 12.01	7 N Nitrogen 14.007	8 O Oxygen 15.999	9 F Fluorine 18.99840323	10 Ne Neon 20.1797
11 Na Sodium 22.98976928	12 Mg Magnesium 24.305	13 Al Aluminum 26.9815385	14 Si Silicon 28.085	15 P Phosphorus 30.973761998	16 S Sulfur 32.06	17 Cl Chlorine 35.45	18 Ar Argon 39.948														
19 K Potassium 39.0983	20 Ca Calcium 40.078	21 Sc Scandium 44.955912	22 Ti Titanium 47.867	23 V Vanadium 50.9415	24 Cr Chromium 51.9961	25 Mn Manganese 54.938044	26 Fe Iron 55.845	27 Co Cobalt 58.933194	28 Ni Nickel 58.6934	29 Cu Copper 63.546	30 Zn Zinc 65.38	31 Ga Gallium 69.723	32 Ge Germanium 72.63	33 As Arsenic 74.921595	34 Se Selenium 78.971	35 Br Bromine 79.904	36 Kr Krypton 83.798				
37 Rb Rubidium 85.468	38 Sr Strontium 87.62	39 Y Yttrium 88.90584	40 Zr Zirconium 91.224	41 Nb Niobium 92.90637	42 Mo Molybdenum 95.95	43 Tc Technetium (98)	44 Ru Ruthenium 101.07	45 Rh Rhodium 102.90550	46 Pd Palladium 106.42	47 Ag Silver 107.8682	48 Cd Cadmium 112.414	49 In Indium 114.818	50 Sn Tin 118.710	51 Sb Antimony 121.757	52 Te Tellurium 127.60	53 I Iodine 126.90447	54 Xe Xenon 131.29				
55 Cs Cesium 132.90545196	56 Ba Barium 137.327	57 - 71 Lanthanoids	72 Hf Hafnium 178.49	73 Ta Tantalum 180.94788	74 W Tungsten 183.84	75 Re Rhenium 186.207	76 Os Osmium 190.23	77 Ir Iridium 192.222	78 Pt Platinum 195.084	79 Au Gold 196.966569	80 Hg Mercury 200.59	81 Tl Thallium 204.38	82 Pb Lead 207.2	83 Bi Bismuth 208.9804	84 Po Polonium (209)	85 At Astatine (210)	86 Rn Radon (222)				
87 Fr Francium (223)	88 Ra Radium (226)	89 - 103 Actinoids	104 Rf Rutherfordium (261)	105 Db Dubnium (262)	106 Sg Seaborgium (266)	107 Bh Bohrium (264)	108 Hs Hassium (265)	109 Mt Meitnerium (268)	110 Ds Darmstadtium (271)	111 Rg Roentgenium (272)	112 Cn Copernicium (285)	113 Nh Nihonium (284)	114 Fl Flerovium (289)	115 Mc Moscovium (288)	116 Lv Livermorium (293)	117 Ts Tennessine (294)	118 Og Oganesson (294)				

How many possible combinations of elements?

2 elements → 5 253

3 elements → 176 851

4 elements → 4 421 275

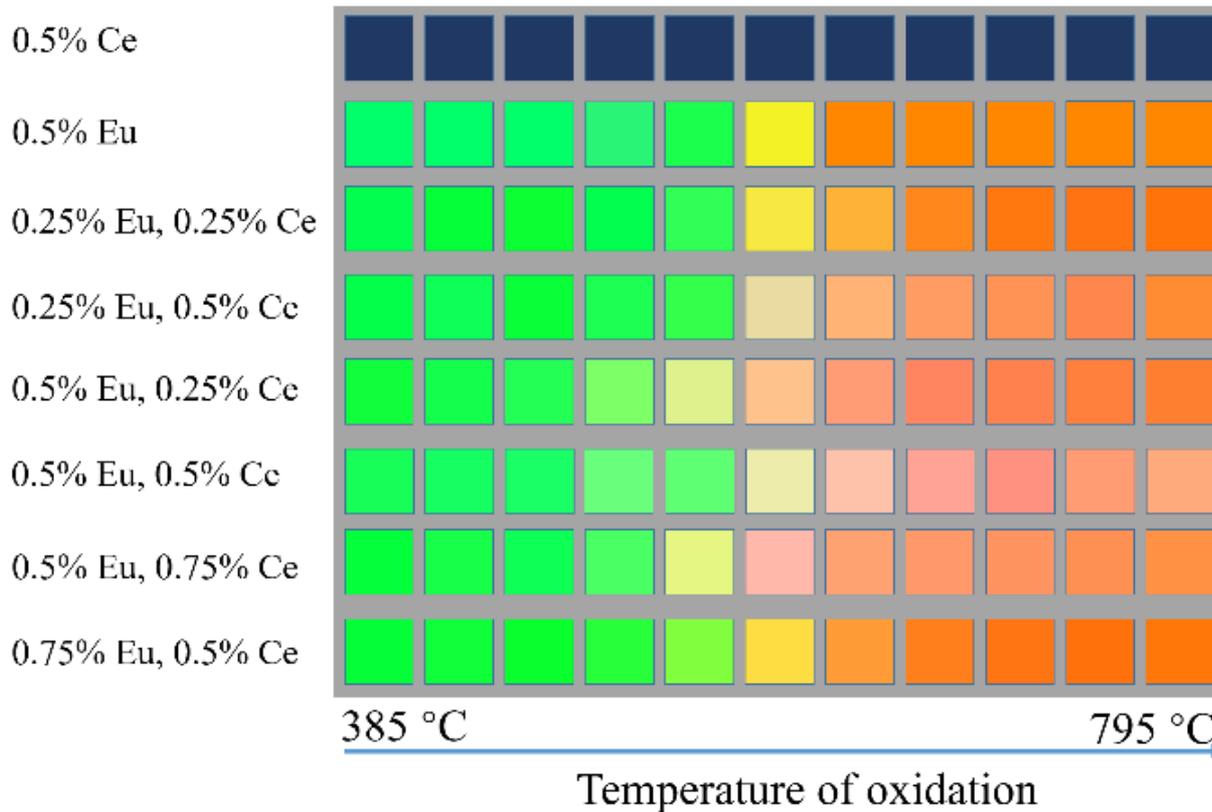
...

57 La Lanthanum 138.9047	58 Ce Cerium 140.16	59 Pr Praseodymium 140.90766	60 Nd Neodymium 144.242	61 Pm Promethium (145)	62 Sm Samarium 150.36	63 Eu Europium 151.964	64 Gd Gadolinium 157.25	65 Tb Terbium 158.92535	66 Dy Dysprosium 162.50	67 Ho Holmium 164.93033	68 Er Erbium 167.259	69 Tm Thulium 168.93422	70 Yb Ytterbium 173.054	71 Lu Lutetium 174.967
89 Ac Actinium 227	90 Th Thorium 232.0377	91 Pa Protactinium 231.03688	92 U Uranium 238.02891	93 Np Neptunium (237)	94 Pu Plutonium (244)	95 Am Americium (243)	96 Cm Curium (247)	97 Bk Berkelium (247)	98 Cf Californium (251)	99 Es Einsteinium (252)	100 Fm Fermium (257)	101 Md Mendelevium (258)	102 No Nobelium (259)	103 Lr Lawrencium (260)

Different stoichiometries, structures of organic molecules,

Number of combinations > Number of atoms in universe

88 samples prepared from the eight synthesized samples by oxidation of the dopants under a gradient of temperature



Colors simulated using the CIE coordinates calculated from the emission spectra of each sample.

❖ Trichromatic emission : three emission centers

