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INFRARED SPECTROSCOPY OF RUTHENIUM TETROXIDE AND HIGH-RESOLUTION ANALYSIS OF THE ν_3 BAND

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INTRODUCTION

After the Fukushima accident, it became essential to have a way to monitor in real time the evolution of a nuclear reactor during a severe accident, in order to react efficiently and minimize the industrial, ecological and health consequences of the accident. Ruthenium is a low volatile fission product but in case of the rupture of the vessel lower head by the molten corium, the air entering into the vessel oxidizes Ru into a gaseous form: RuO_4 . To monitor the presence of RuO_4 allows making a diagnosis of the core degradation and quantifying the potential release into the atmosphere. RuO_4 is of prime importance since it can have a significant radiological impact. Here, we present the study of the feasibility of the monitoring by an optical measurement.

MATERIAL AND METHODS

RuO_4 is very reactive: Requires a synthesis and analytical process fully passivated.

Synthesis process

- Alkaline fusion of Ruthenium metal with potassium hydroxide
- Oxidation with potassium permanganate
- Trapping RuO_4 gas in dry ice



Measuring system

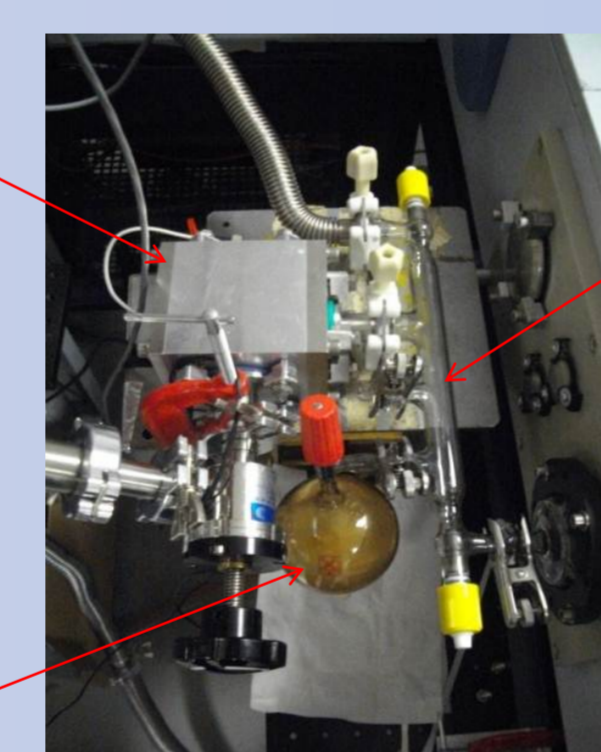
Measuring cell and specific pressure sensors, fully passivated.

- Glass cell (124 mm)
- ZnSe windows treated AR reflexion
- Kalrez seals and teflon valves
- Pressure sensors developed at the French SOLEIL synchrotron facility (L. Lago)
- whose surfaces exposed to the gas were passivated (res: 0.02 mbar)

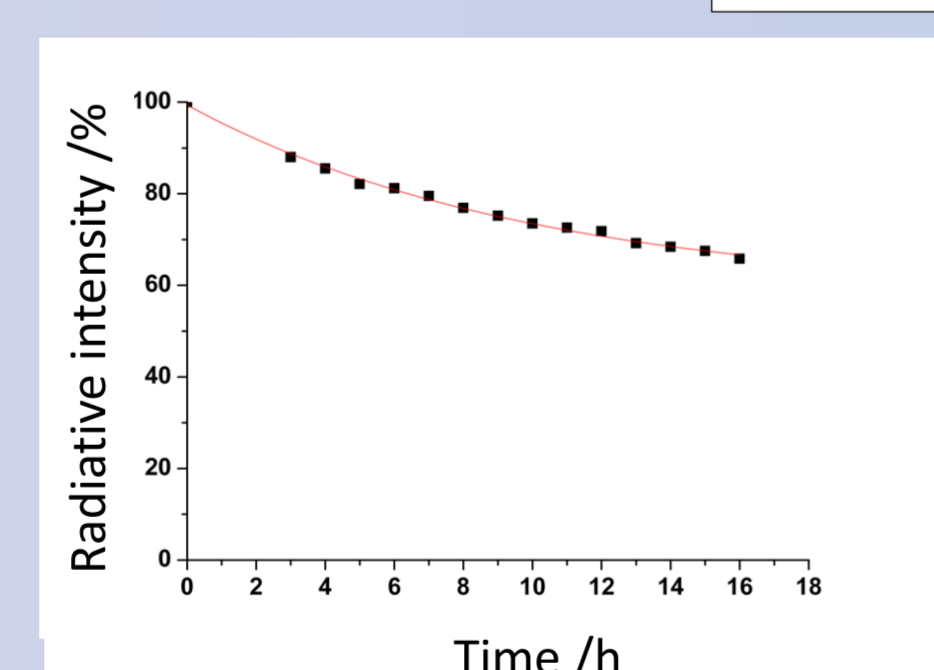


Pressure sensors

RuO₄ bulb



Ramp to the cell



BUT

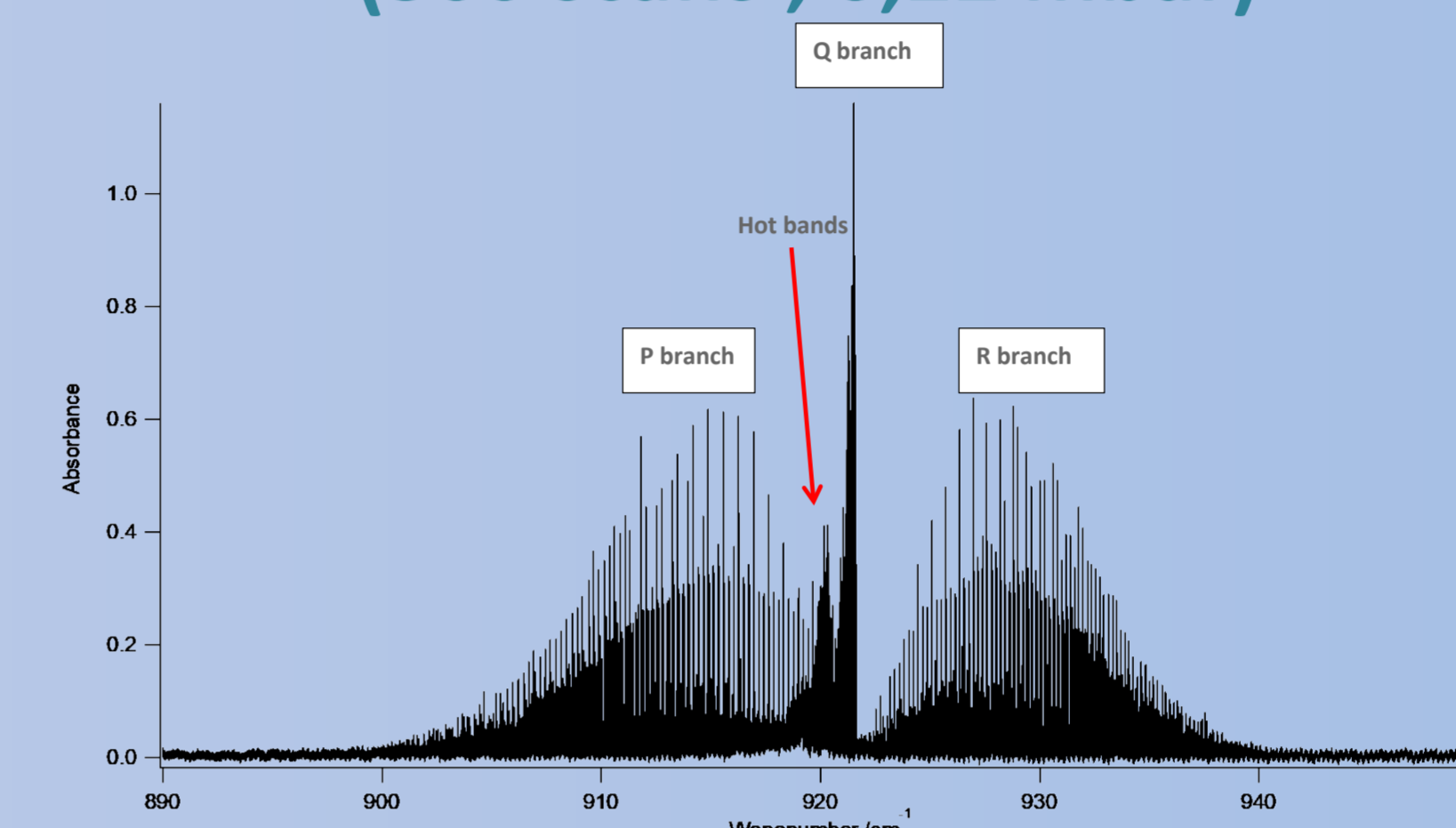
RuO_4 ν_3 intensity decreases over time

PERSPECTIVES

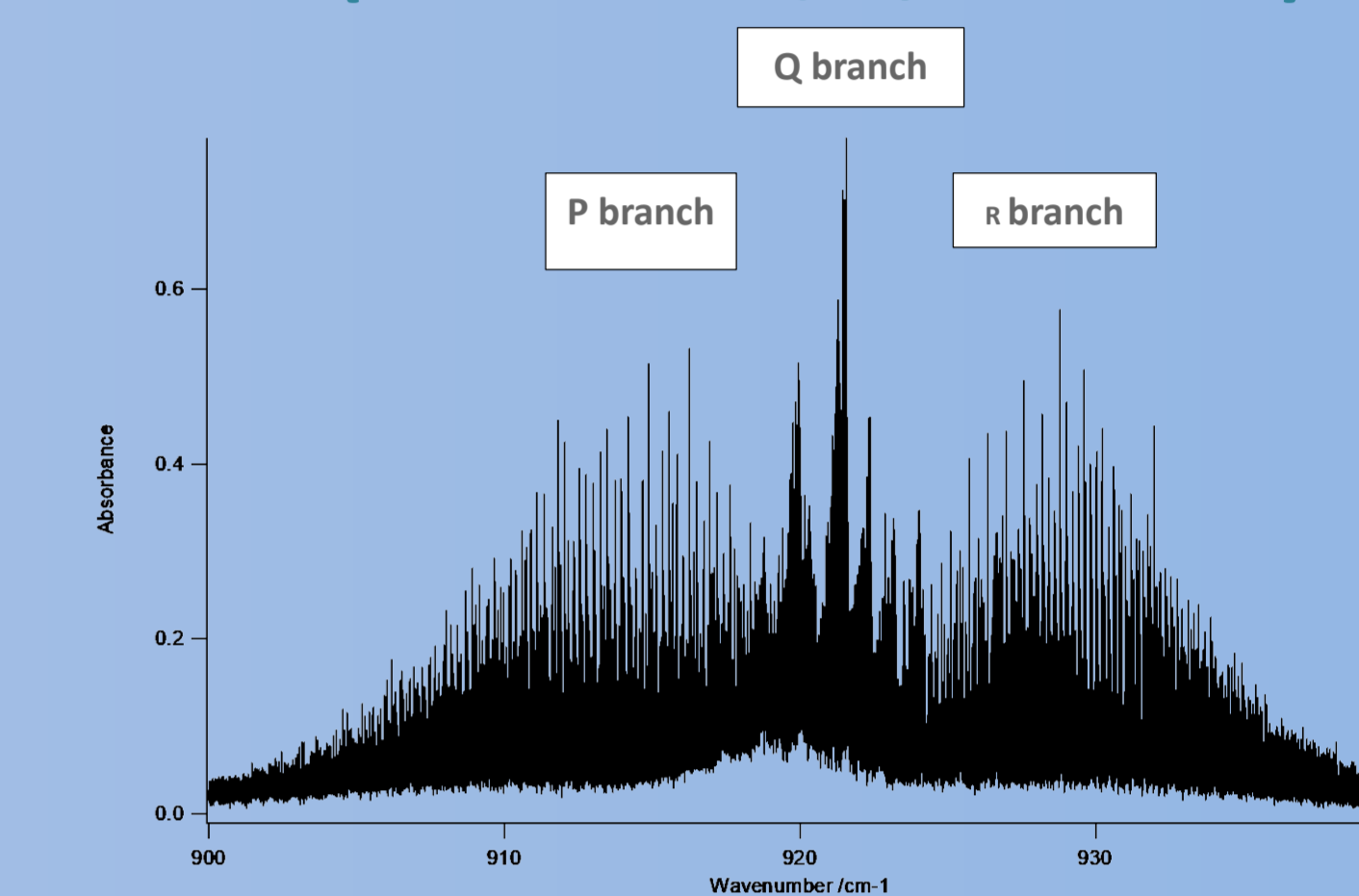
- Analysis by high-resolution Raman spectroscopy to determine the parameters of the ν_1 vibration mode (**In progress**)
- An analysis by high-resolution in the far infrared, to study the ν_2/ν_4 bending dyad (**In preparation**)
 - The determination of the ν_1, ν_2, ν_3 and ν_4 harmonic wavenumbers values => RuO_4 equilibrium bond length.
 - It would allow the study of higher harmonics of the RuO_4 bending modes.
 - It would allow the identification of weak forbidden lines that permit to fit ground state parameters.
- Determination of the spectral broadening: Simulation of RuO_4 dilute in the air: quantification of the release
- Development of a new sensor based on an optical measurement (LIDAR, CES,...)

Analysis by high resolution FTIR spectroscopy (SOLEIL synchrotron, AILES beamline)

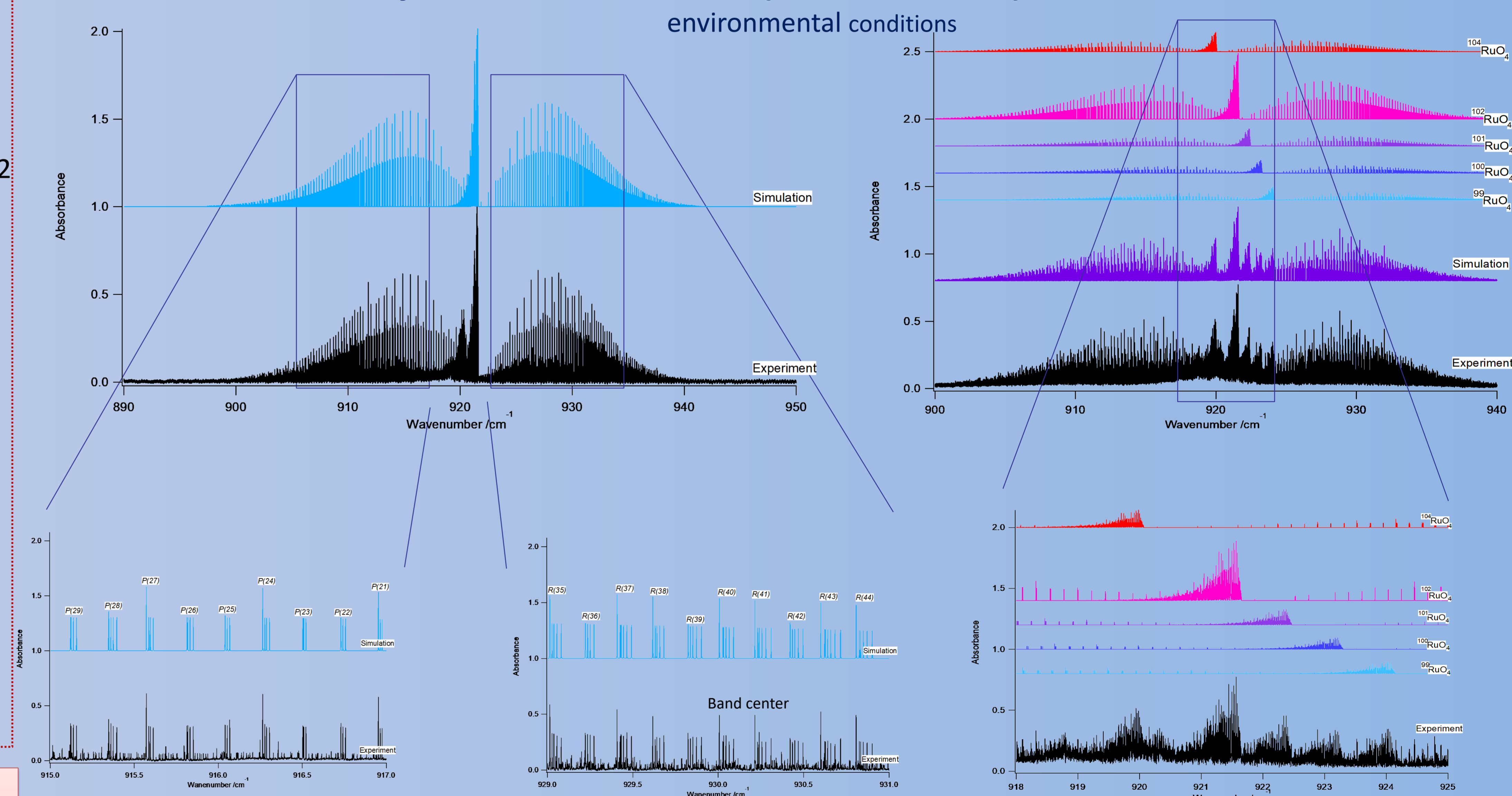
$^{102}\text{RuO}_4$ monoisotopic
(300 scans ; 0,22 mbar)



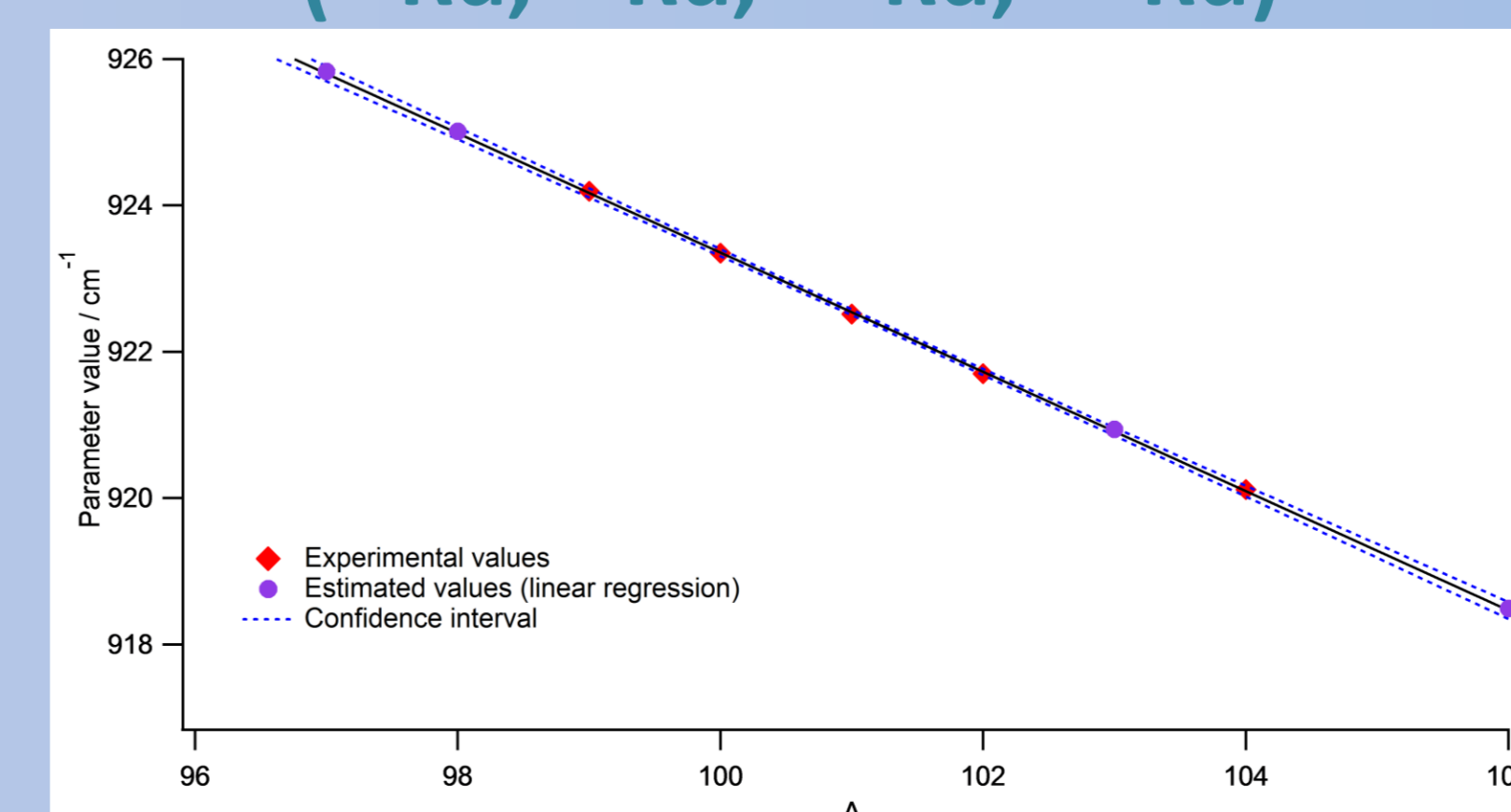
RuO_4 in natural abundance
(680 scans ; 0,72 mbar)



Simulation by tensorial formalism (XTDS, SPVIEW) => Reference spectra whatever the environmental conditions



Extrapolation of parameters for minor isotopologues ($^{97}\text{Ru}, ^{98}\text{Ru}, ^{103}\text{Ru}, ^{106}\text{Ru}$)



Improving data

