



**Fiber-coupled Raman probe**  
**designed for H<sub>2</sub>-risk management during a severe accident.**  
*Proof-of-Concept for remote gas monitoring in nuclear containments*



Session #8 – Severe Accident Monitoring



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# Summary

- 1. The H<sub>2</sub>/MCCI risk and management strategies**
- 2. MITHYGENE Project : rationale, structure and chronology**
- 3. Raman spectrometry (basics)**
- 4. MITHYGENE #1 (2013-2018) : Feedback**
- 5. MITHYGENE #2 (2019-2022) : new probe**
- 6. Gas qualification (CNRS-ICARE)**
- 7. Radiation qualification (IRSN)**
- 8. Conclusions & perspectives**

# Severe Accident (SA) in light water reactors (1/2)

Oxydation of fuel rods at high  $T^\circ$  (loss of coolant)

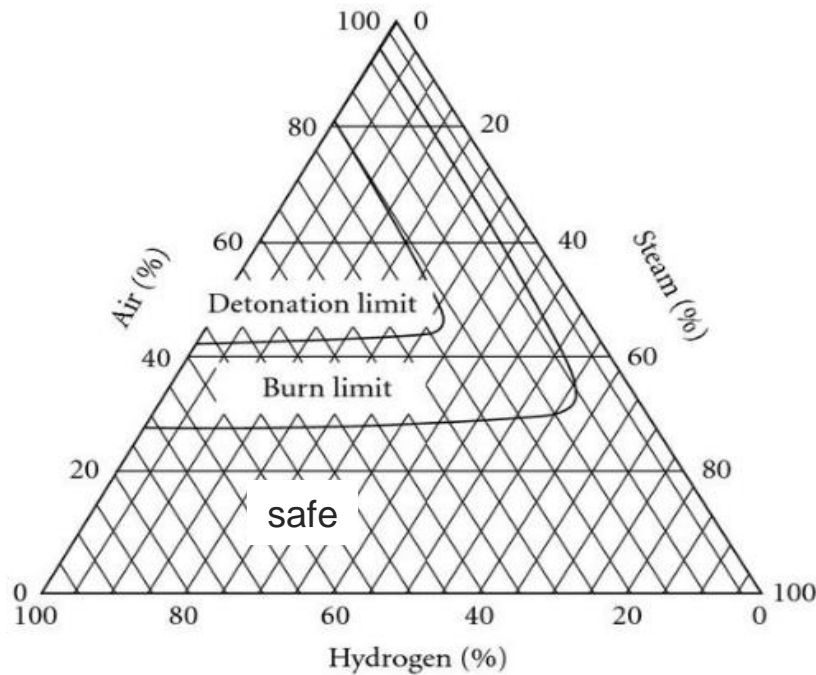


Release of  $H_2$  gas within the containment building and annulus

Molten Corium-Concrete Interaction (MCCI)



Additional release of  $H_2$  and CO gas (CO = MCCI tracer)



TERNARY DIAGRAM (SHAPIRO-MOFFETTE):  
safe, deflagration & detonation situations

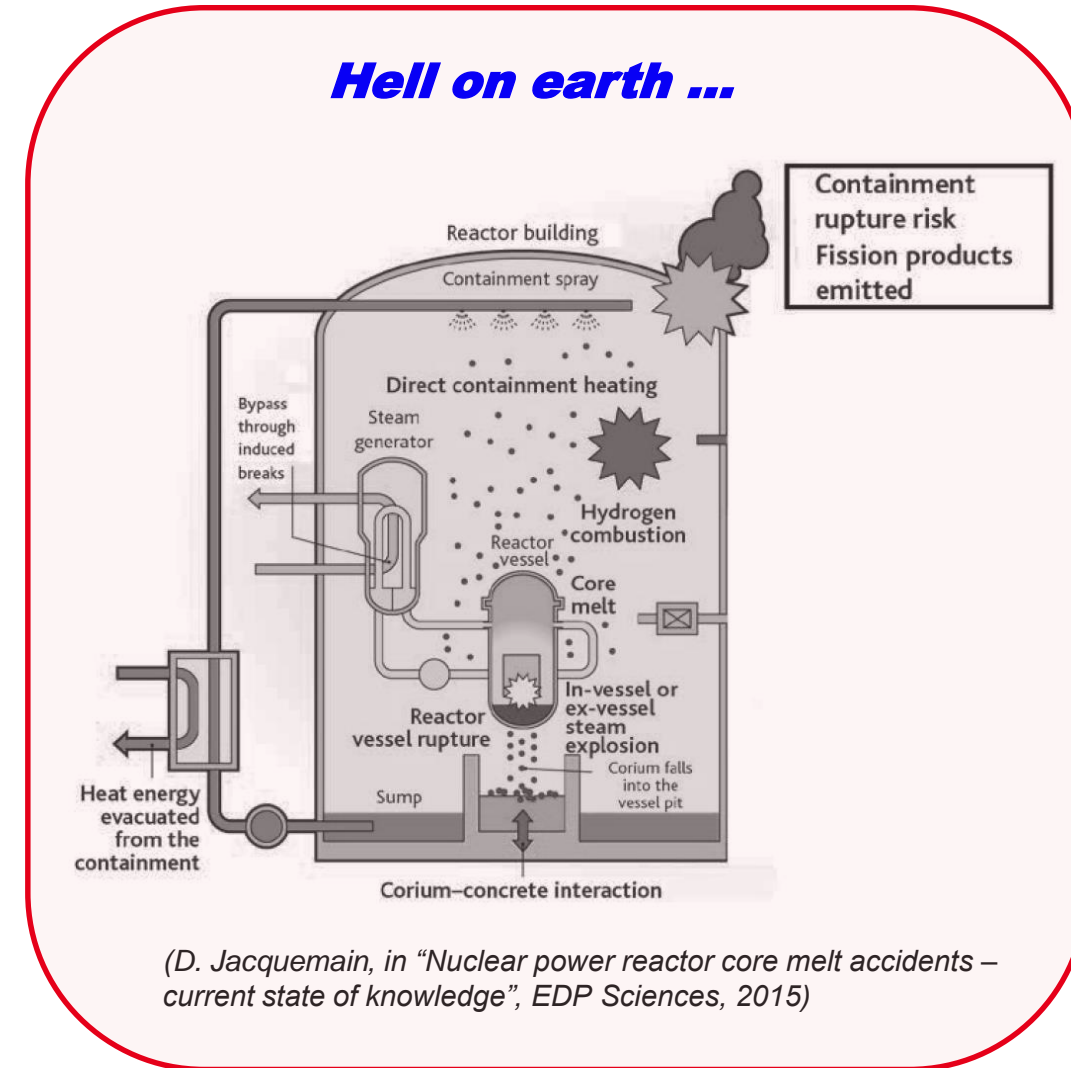


# Severe Accident (SA) in light water reactors (2/2)

Loss-of-Coolant Accident leading to H<sub>2</sub> release and core meltdown

Environmental specifications (french RCC standard)

- Pressure : **9 bar (abs)**
- Peak temperature : **170°C** (during several hours)
- Dose / Dose rate (air) : **2 MGy / 1 kGy/h** (air)
- Saturation level of water vapor **RH = 100 %** (during several hours)
- Presence of **aerosols**
  - soot due to combustion,
  - emission of fission products, metallic & nuclear fuel residuals
- **Shocks/vibrations** (Design Basis Earthquake – DBE)
- Design basis and SA-qualified hardware :
  - Standalone during entire operational lifetime  
(minimum 40 years, probably more)
  - MTTF > several months after the onset of the accident.



# H<sub>2</sub>-risk management strategies (1/2)

H<sub>2</sub> Mitigation strategy → NPP-design dependent

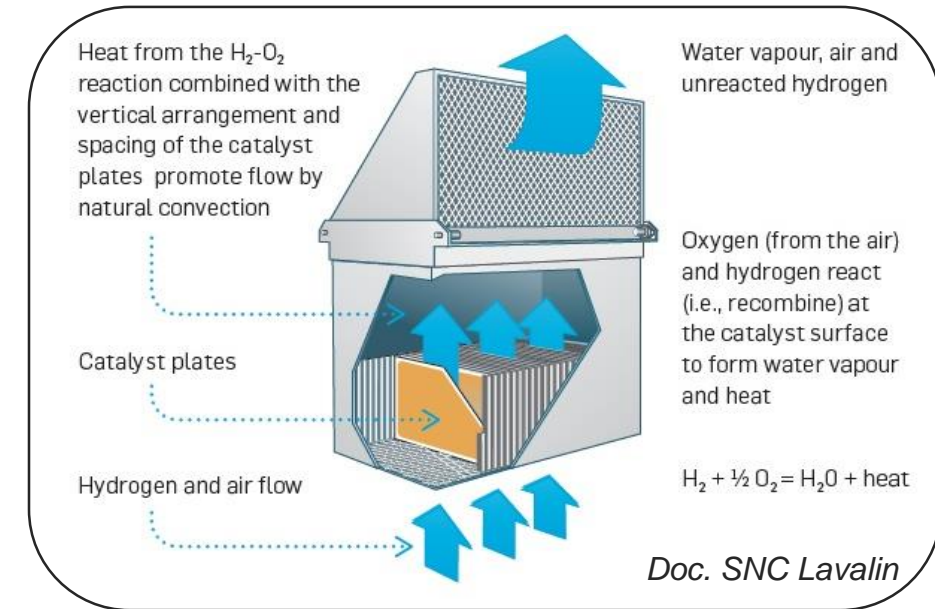
Combines the use of safety devices (PARs, etc.)  
with SAMG (*Severe Accident Management Guidelines*)

## Principle of Passive Autocatalytic Recombiners (PARs)

- Exothermic reaction process (*self-started, no power required*),
- Presence of H<sub>2</sub> inferred by monitoring temperature (*thermocouple, TC*) .

## Limitations

- The H<sub>2</sub> recombination rate may be much lower than the generation rate : **partial mitigation**,
- Local increase in T° → convection flows,
- Poisoning by other gases released during the accident (CO, I<sub>2</sub>, CH<sub>3</sub>I, etc.) or by soot,
- Few PARS are actually equipped with TC (on polar crane),
- **PARs do not provide assessment of H<sub>2</sub>-risk.**



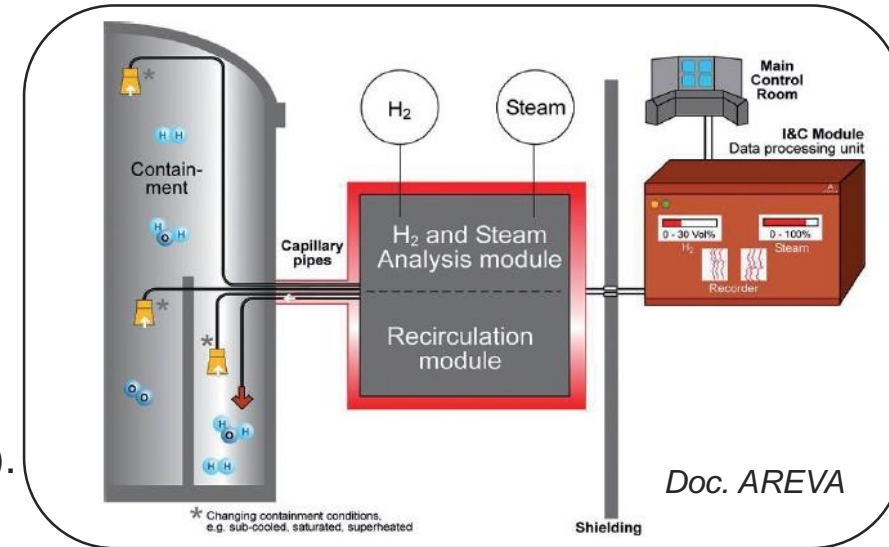
# H<sub>2</sub>-risk management strategies (2/2)

## Principle of HERMETIS system (ex AREVA, now Framatome)

- Overheated sampling pipes transport gas samples out of the concrete barrier into a monitoring module installed outside
- Superheated gas microsampling (capillary pipes) at several locations within the containment,
- Selective monitoring of H<sub>2</sub> and H<sub>2</sub>O,
- Capability for CO and O<sub>2</sub> (*dedicated to BWR*)

## Limitations

- **French safety doctrine precludes sampling through the containment wall**
- Gas segregation onto pipe walls → **biased measurement** H<sub>2</sub>O ↔ O<sub>2</sub>, H<sub>2</sub>,
- Gas transit time → **delayed response**,
- Sampling over large volumes → local heterogeneities are averaged out,
- Does not provide N<sub>2</sub> and CO<sub>2</sub> monitoring,
- Requires local power supply → failure in case of electrical shutdown (Fukushima).



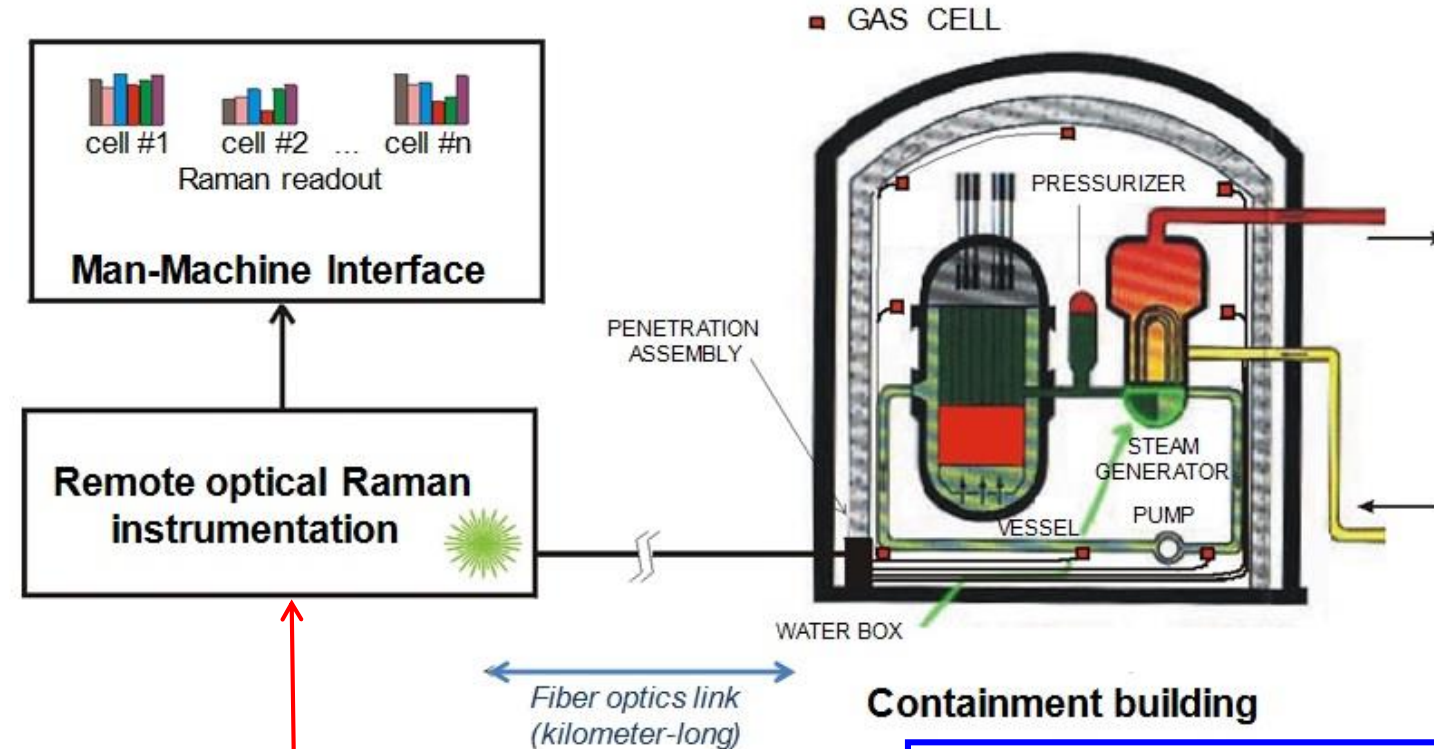
Alternative solution = *in situ* monitoring of gases within the containment  
**Rationale for Mithygene Project**

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# MITHYGENE Project : rationale



- Monitoring unit placed in safe zone, beyond radiological perimeter
- Battery-powered in case of electric shutdown
- Probe installation according to accidental models performed by IRSN.

 **Emergency power supply**  
out of radiological perimeter

## Containment building

- ✓ 6 targeted molecules :  $H_2$ ,  $O_2$ ,  $N_2$ ,  $H_2O$  ( $H_2$ -risk),  
 $CO$ ,  $CO_2$  (MCCI),
- ✓ *In situ* (no sampling), local and multipoint monitoring,
- ✓ ATEX compatible,
- ✓ Response time < 10 minutes (SAMG),
- ✓ High selectivity, no cross-sensitivity (e.g. vs  $H_2O$ )
- ✓ Absolute monitoring of partial pressures (internal Raman reference)



# MITHYGENE Project : structure & chronology

MITHYGENE #1 (2013-2018)



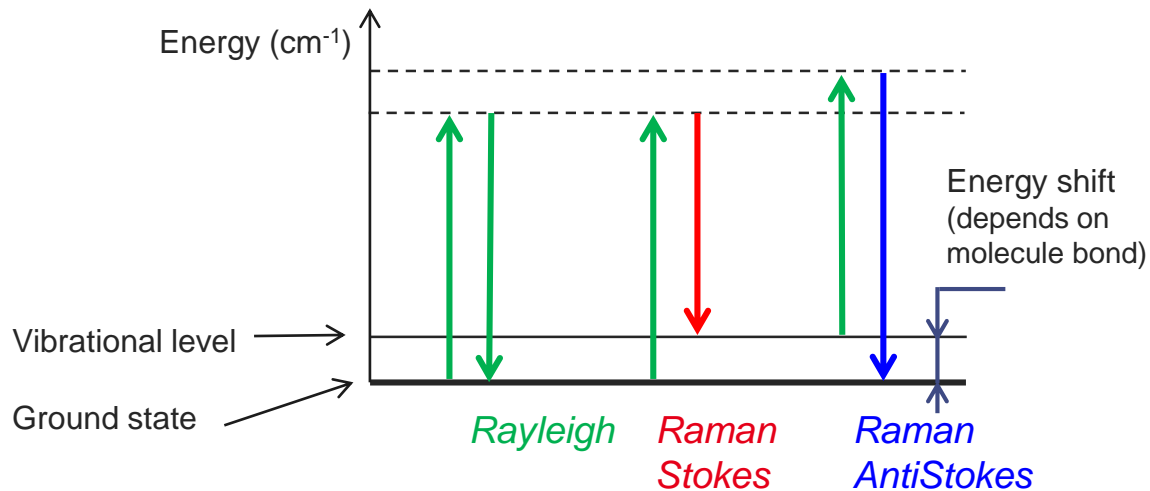
MITHYGENE #2 (2019-2022)  
Tripartite Project Extension



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# Raman spectrometry (basics, 1/3)



## Raman cross-sections

$$\frac{d\sigma}{d\Omega} \approx 1/\lambda^4$$

$$\left. \frac{d\sigma}{d\Omega} \right|_{N_2} = 1.45 \cdot 10^{-31} \text{ cm}^2/\text{Sr}$$

$$\left. \frac{d\sigma}{d\Omega} \right|_{H_2} = 3.6 \cdot 10^{-31} \text{ cm}^2/\text{Sr}$$

(@640 nm)

## Raman features

- ✓ Simplicity (1 laser, 1 spectrometer)
- ✓ Chemical selectivity
- ✓ FWHM < 12 nm → selectivity (discrimination CO/N<sub>2</sub> and CO<sub>2</sub>/O<sub>2</sub>)  
→ Possible use of compact, low-cost spectrometer
- ✓ No interference with water (absorption)
- ✓ Laser emitting in the red domain (@730 – 750 nm)  
→ Detection with PMs is (still) possible

## General law for Raman signals with gas

$$S \cdot T = K \cdot P_{\text{Laser}} \cdot \left( \frac{d\sigma}{d\Omega} \right) \cdot \Delta\Omega \cdot L \cdot P \cdot \Delta t$$

Temperature →  $\left( \frac{d\sigma}{d\Omega} \right)$

Solid angle →  $\Delta\Omega$

Absolute pressure →  $P$

Raman signal ←  $S \cdot T$

Laser power ←  $P_{\text{Laser}}$

Interaction length ←  $L$

Integration time ←  $\Delta t$

# Raman spectrometry (basics, 2/3)



Gas	$\sigma/\sigma_{N_2}$	Depolarization ratio $\rho$	$\Delta\nu$ (cm <sup>-1</sup> )	$\lambda$ Raman (nm) @ 730 nm	$\lambda$ Raman (nm) @ 750 nm
H <sub>2</sub>	2.4	~1.2 %	4156	1048	1089
	2.4	75 %	587	763	785
H <sub>2</sub> O	3.2	~0.03 %	3657	996	1033
N <sub>2</sub>	1	~1 %	2331	880	909
CO	1	~0,6 %	2145	866	894
O <sub>2</sub>	1.2	~3 %	1556	824	849
CO <sub>2</sub>	1.3	~4 % to 5 %	1285 and 1388	806 and 812	830 and 837

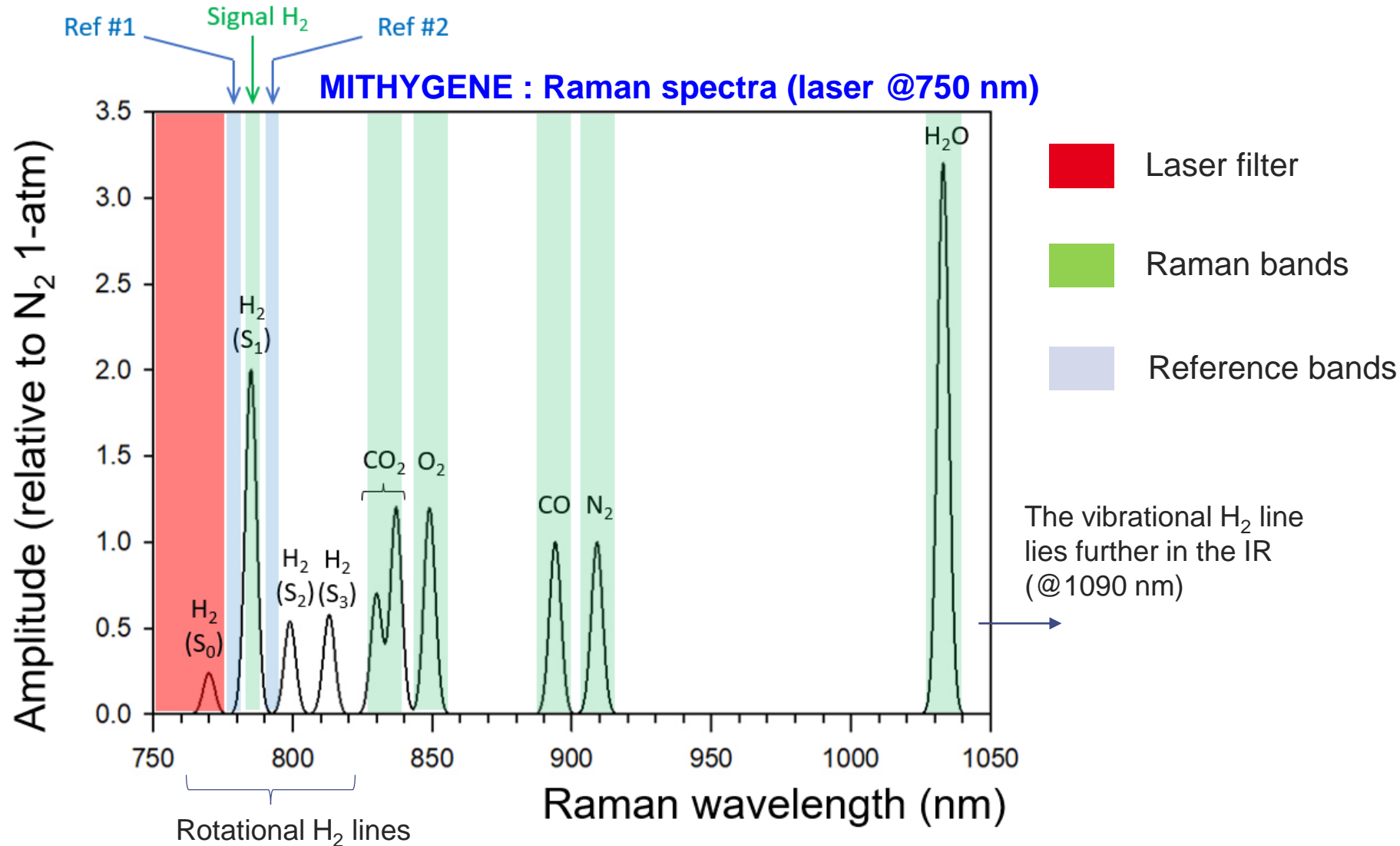
Shift : 186 cm<sup>-1</sup>

Shift : 168 cm<sup>-1</sup>

## To learn more ...

S. Magne, S. Nehr, X. Buet, A. Bentaib, E. Porcheron, R. Grosseuvres, E. Studer, R. Scarpa, D. Abdo, J-L. Widloecher, O. Norvez, N. Chaumeix, J. Dhote, M. Freyssinier, A. Ruffien-Ciszak, *In situ* gas monitoring by Fiber-Coupled Raman Spectrometry for H<sub>2</sub>-risk management in nuclear containment during a severe nuclear accident, IEEE Trans. Nucl. Sci. 67 (4), 2020, pp. 617-624

# Raman spectrometry (basics, 3/3)



- ▶ Laser CW (@750 nm, 3.5W)
- ▶ Spectrometer Shamrock
- ▶ CCD 2D Si IDUS416 (Andor)

*Optional*  
 Lasers (@640nm, @730 nm)  
 Laser modulation driver  
 1x4 coupler (multitrack operation)

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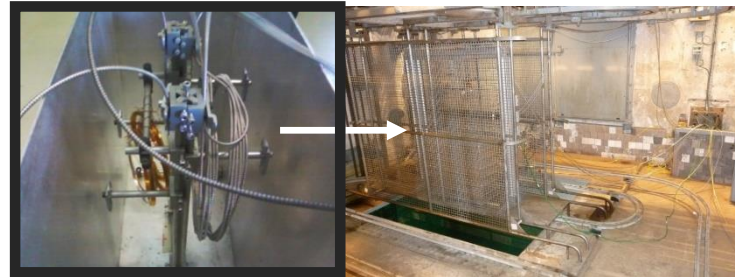
# MITHYGENE #1 Feedback



😊 Test in climatic chamber (CEA)  
[80% RH, 15° – 85°C]



😊 SA thermodynamic conditions  
MISTRA, CEA [4,2 bar, 130°C, 100%RH]

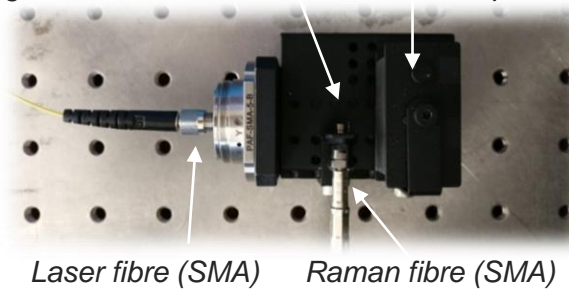


☹ Irradiation test (Poseidon, CEA) [<sup>60</sup>Co, 5 kGy/h, 1.9 MGy]

Raman nuclearized probe (single pass, transverse)

**FOR USE IN AMBIENT AIR**

Light collection at waist      laser dump



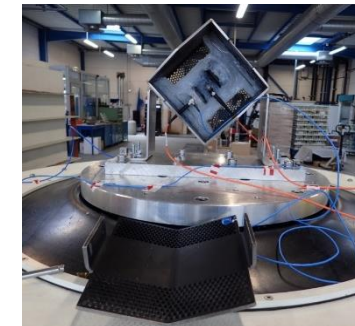
Laser fibre (SMA)      Raman fibre (SMA)



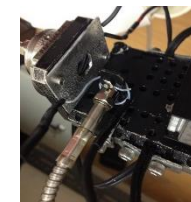
😊 H<sub>2</sub> combustion test (SEXHY, CEA)



😊 Shock test IK07 (LNE)



😊 Vibration test (LNE)  
(NF EN 60028-2-6)



😊 Anti-aerosol  
& antifogging tests  
(CEA, IRSN)

➡ MITHYGENE #2 : Design of a new probe (more efficient) + antifogging/anti-aerosol module and tests

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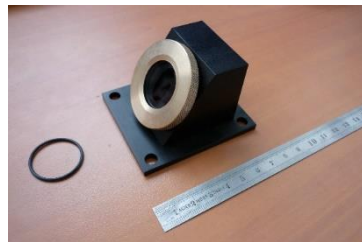
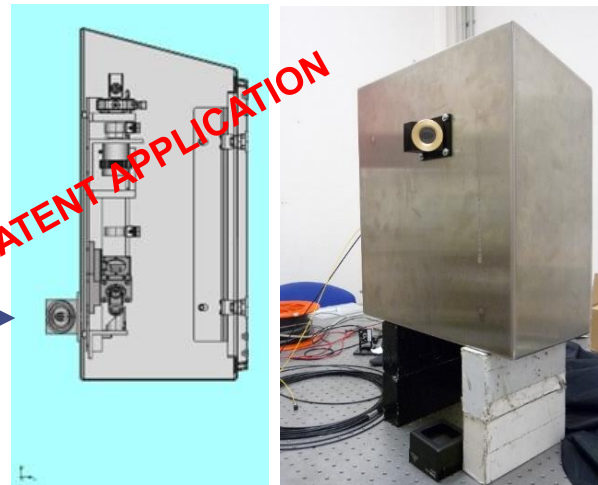


# MITHYGENE #2 : new Raman probe

Single pass **longitudinal** probe  
MITHYGENE-EXTENSION  
(2019-2022)

**IN SEALED ENCLOSURE (IP69K)**

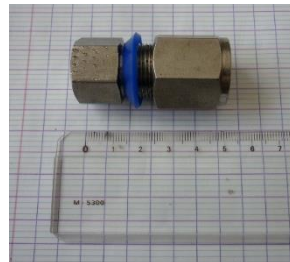
Anti-fogging  
& anti-aerosol module  
will be fixed  
onto probe output



Brewster window  
(laser & Raman collection)

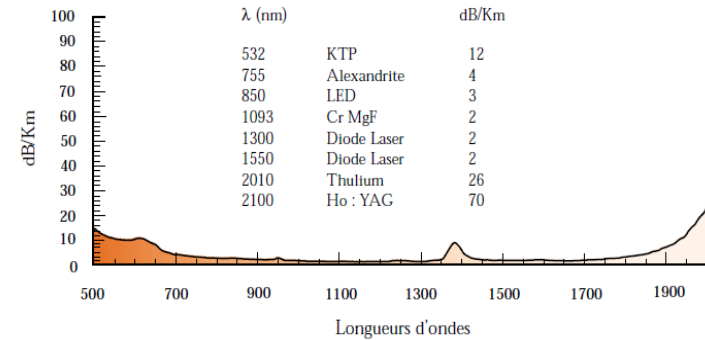


KTRAV (SEDI)  
(fiber optics feedthrough)



Swagelok fitting  
(electrical feedthrough)

Pure silica-core (PSC) fiber (low OH)



Parameter	MITHYGENE Probes	
Interaction mechanism	Transverse (2013-2018)	Longitudinal (2019-2022)
Ø Raman fiber	200 $\mu\text{m}$	600 $\mu\text{m}$
Ø Laser fiber	100 $\mu\text{m}$	400 $\mu\text{m}$
Laser power (unit)	0.7 W	3.5 W
Laser power (gas)	0.5 W	0.6 W
Laser wavelengths	@640 nm @730 nm	@750 nm
Raman efficiency for N <sub>2</sub> detection (0,78 atm, in Cts.s.W)	620	10500 <b>x17</b>

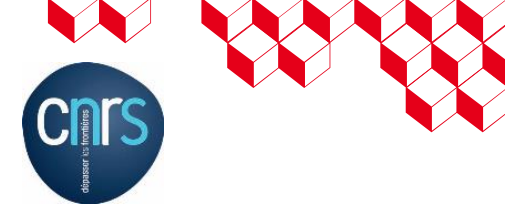
(experimental)

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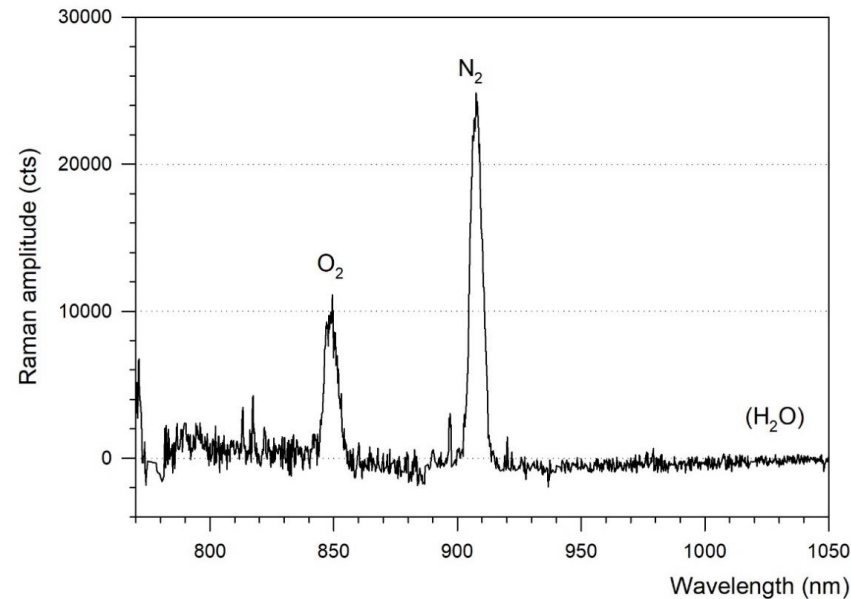
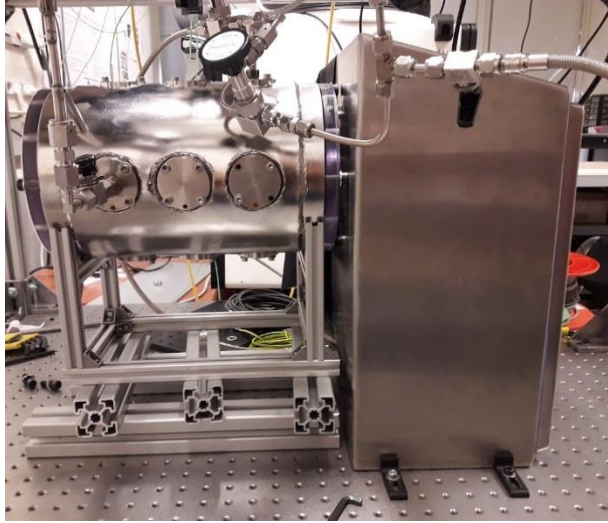


# Gas qualification : H<sub>2</sub>, CO and CO<sub>2</sub> (ICARE)



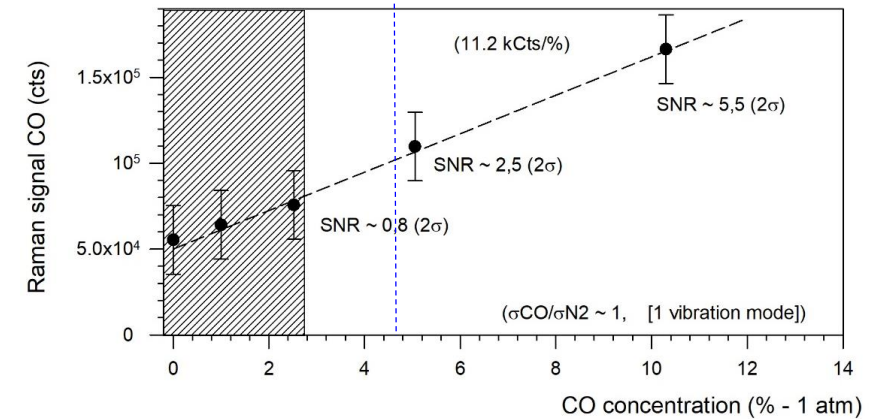
## CO and CO<sub>2</sub> qualification

(SNR = Signal-to-Noise Ratio)

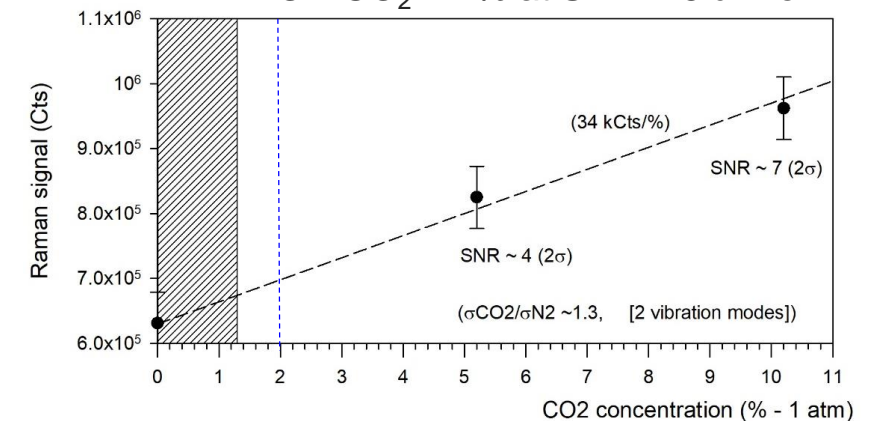


- NC2V cell operated by ICARE,
- Available gases : H<sub>2</sub>, [50% H<sub>2</sub> – 50% N<sub>2</sub>], [50% H<sub>2</sub> - 50% CO], CO<sub>2</sub>
- Homogeneity and pressure control by vacuum pumping,
- Raman probe put aside to NC2V (*Brewster window was removed*), dump placed inside.

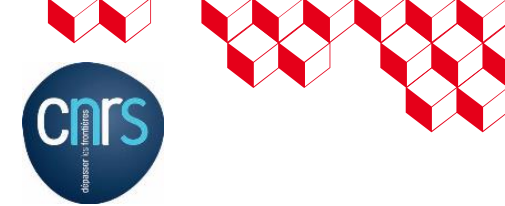
- 1 W laser, 10 minutes,
- LOD CO ~ 4.5% at SNR = 3 à 1- $\sigma$



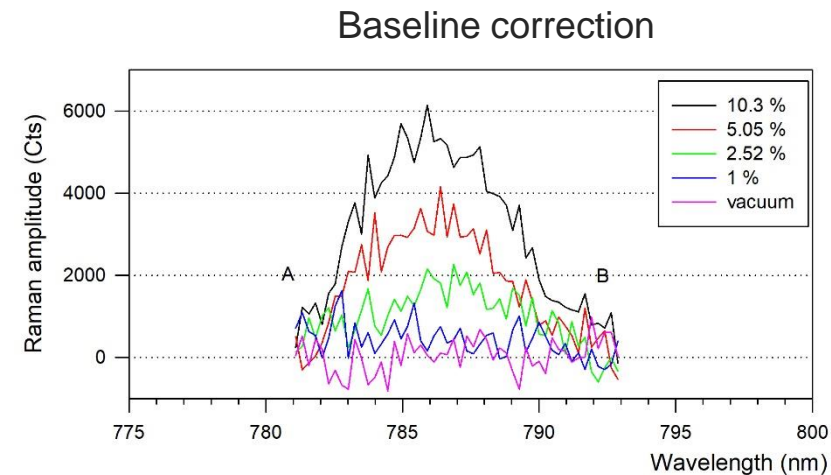
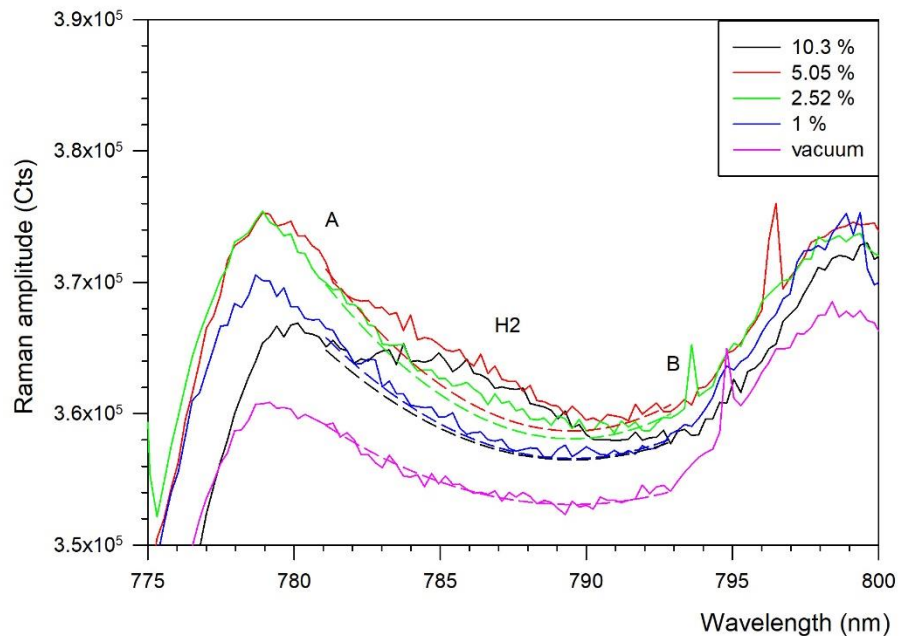
- 1 W laser, 10 minutes,
- LOD CO<sub>2</sub> ~ 2% at SNR = 3 à 1- $\sigma$



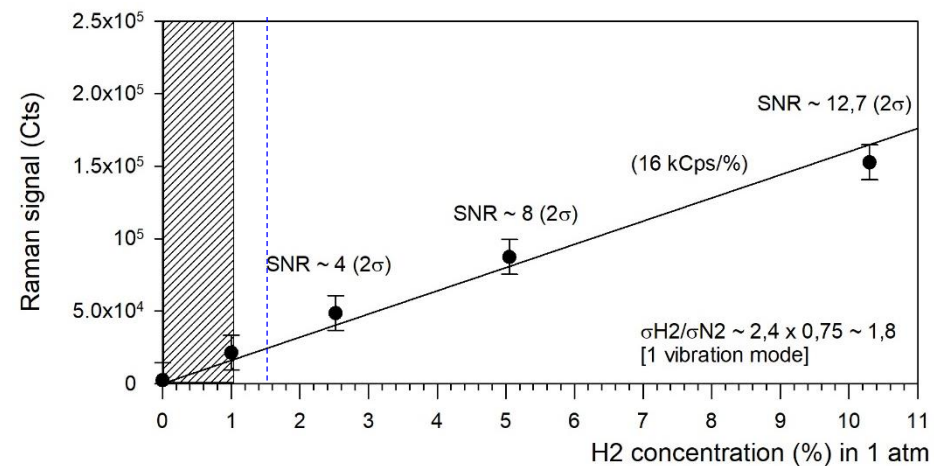
# Gas qualification : H<sub>2</sub>, CO and CO<sub>2</sub> (ICARE)



## H<sub>2</sub> Qualification (baseline correction of rotational bands)



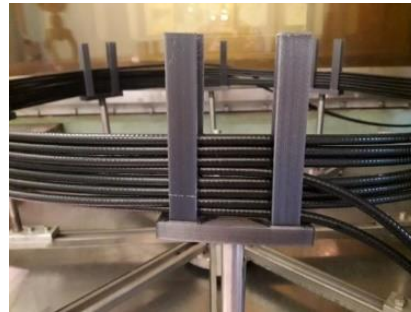
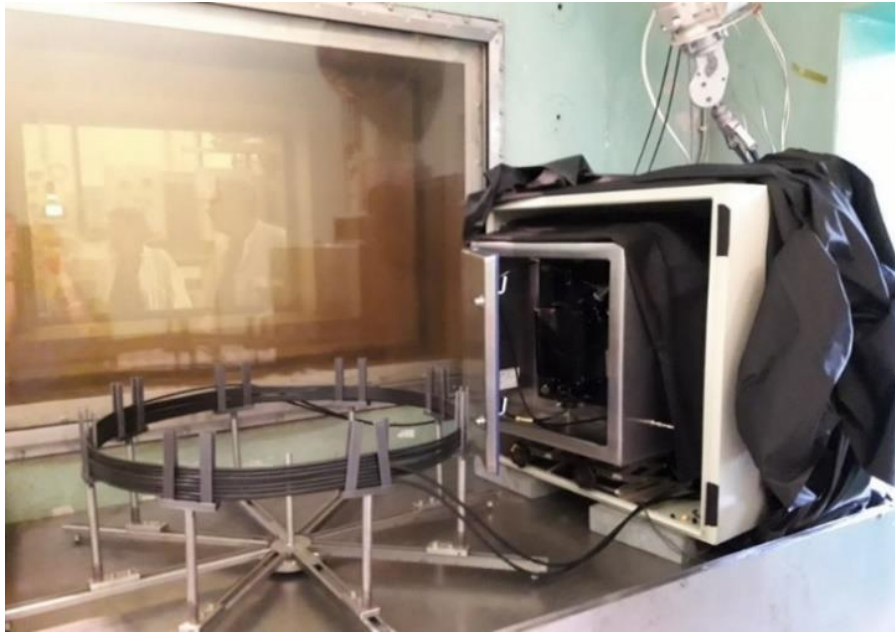
- 1 W laser, 10 minutes,
- LOD H<sub>2</sub> ~ 1,5% at SNR = 3 à 1-σ



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# Radiation qualification (1/4)



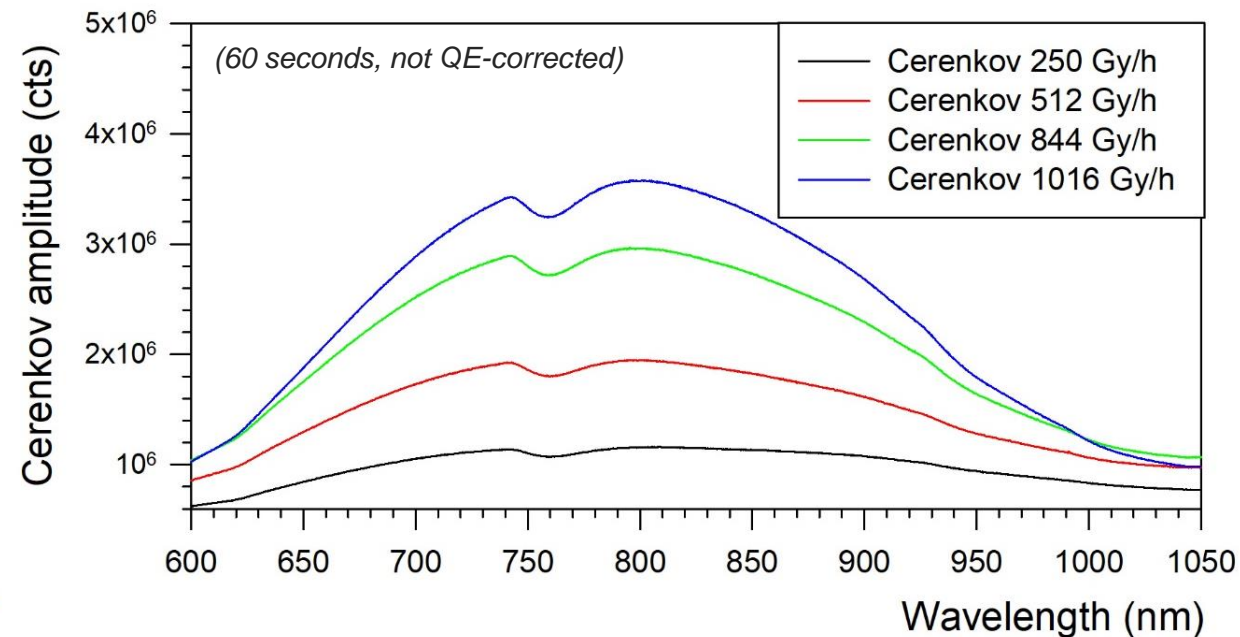
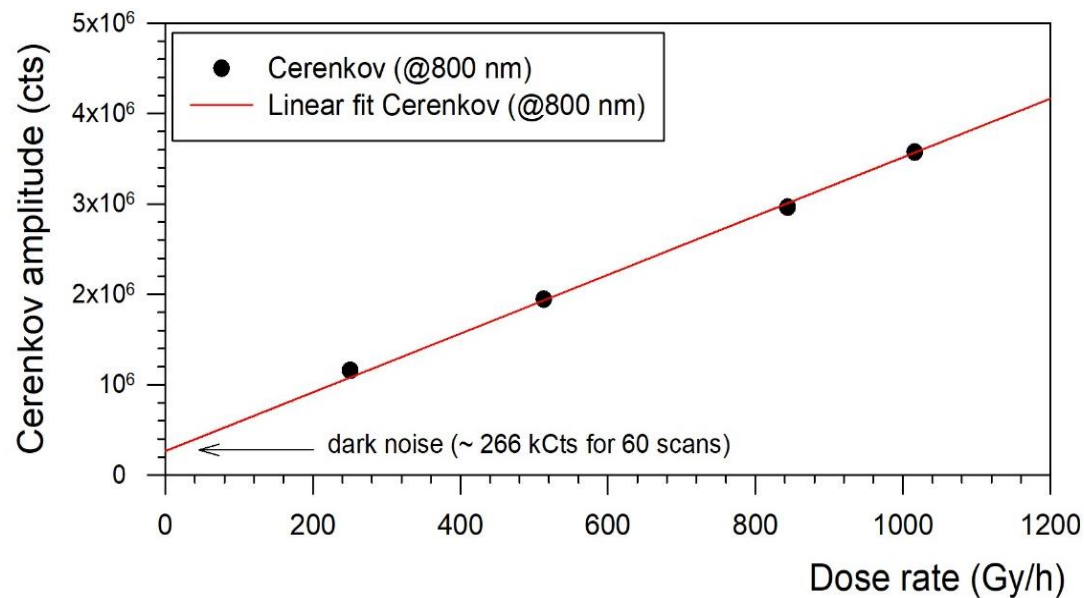
- $^{60}\text{Co}$  sources placed at center of circular isodose,
- Laser and Raman fibers spun around a 61-cm diameter support,
- Total exposed length ~ **10 meters**,
- **4 dose rates: 250 Gy/h, 513 Gy/h, 844 Gy/h and 1016 Gy/h**
- Dose rate on probe ~  $\frac{1}{4}$  dose rate over fibers (228 Gy/h vs 1 kGy/h),
- Laser dump inside inox package (light-protected),
- $T^\circ \sim 30^\circ$  (constant).
- Raman spectra of air and  $\text{H}_2$  (TEDLAR bag).

# Radiation qualification (2/4)

## Influence of Cerenkov light on Raman signals



The Cerenkov effect is highly detrimental to Raman measurements  
(*actually the most detrimental factor*)



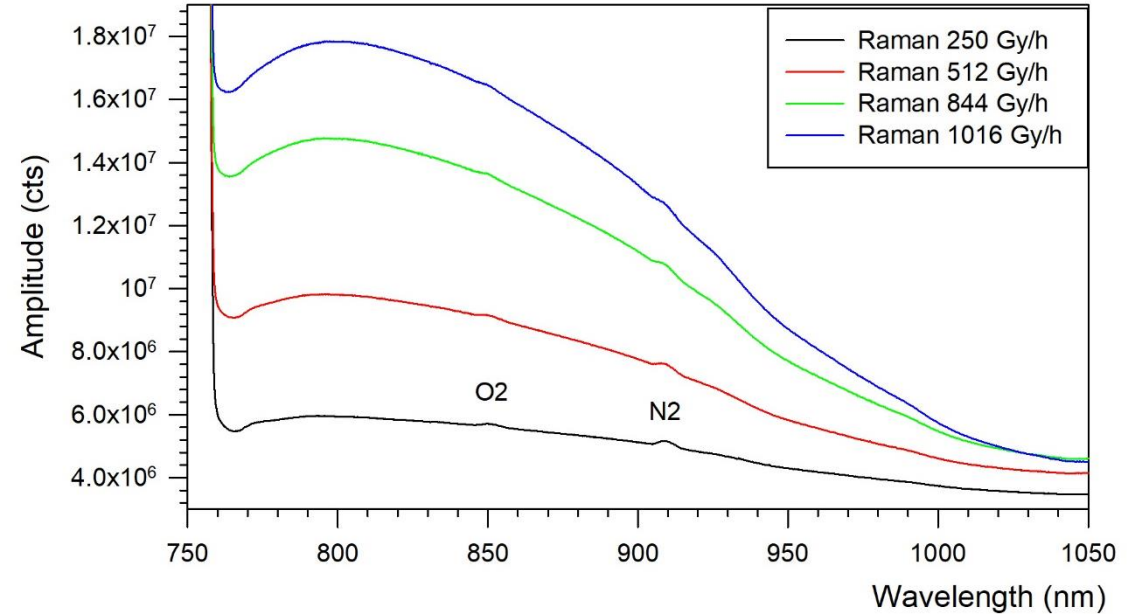
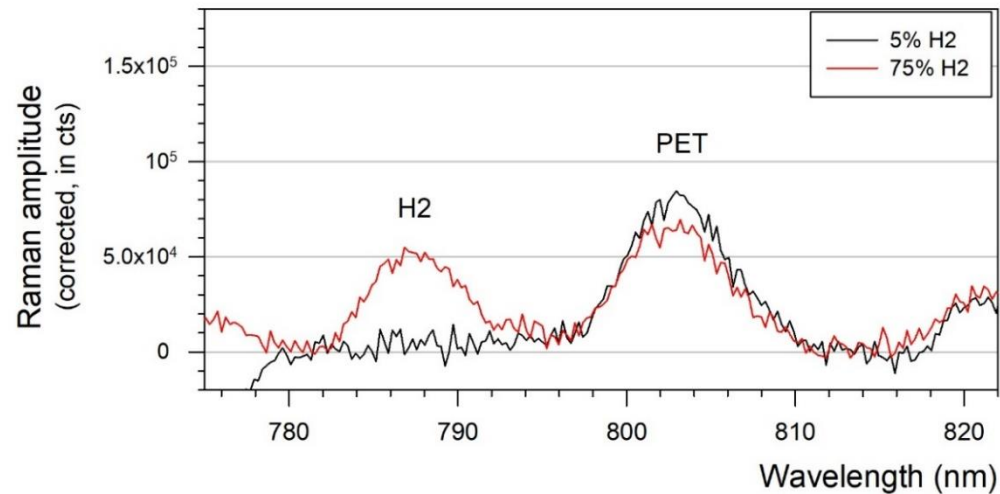
But ..  
added value to the H<sub>2</sub>/MCCI risk management !

→ Real-time multizone (average) dosimetry  
inside the nuclear containment ...

# Radiation qualification (3/4)

## Influence of Cerenkov light on Raman signals

Raman spectra of air  
over Cerenkov light  
(3.5 W, 300 s)



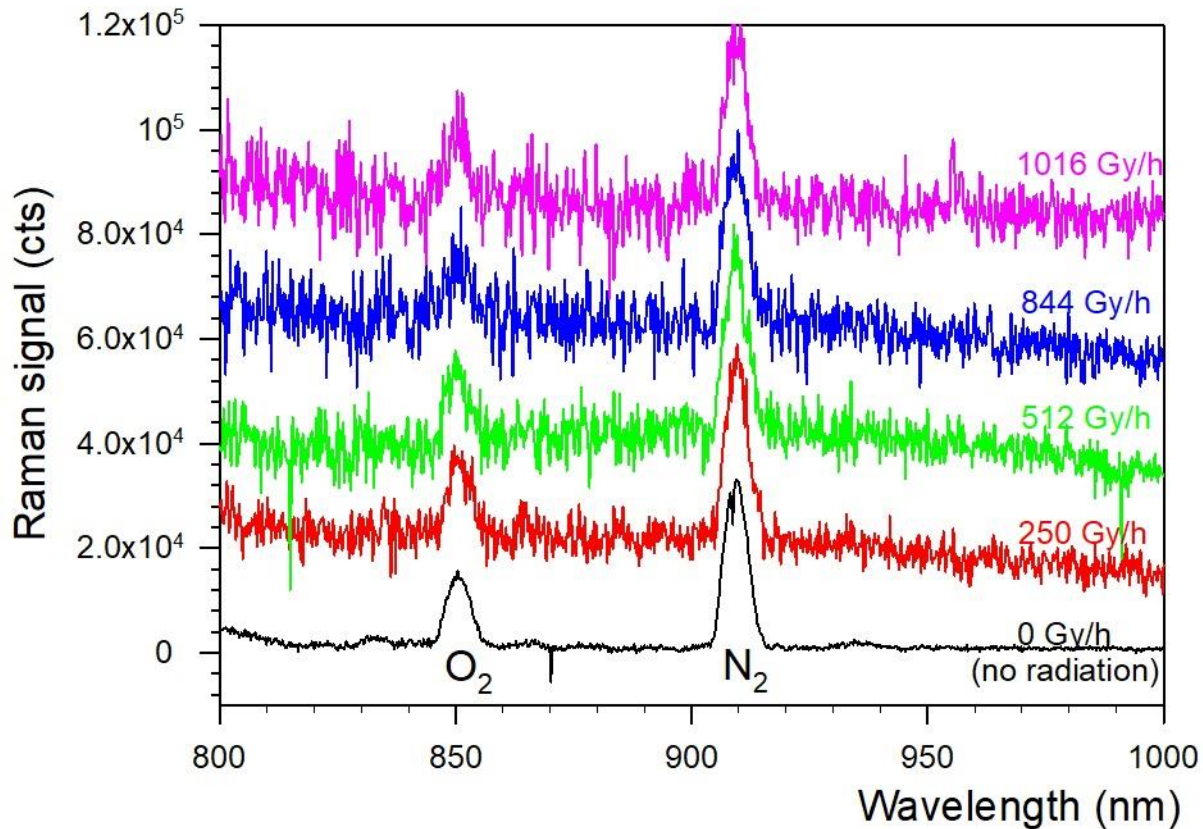
◀ Raman spectra of H<sub>2</sub> (in TEDLAR bag)  
(corrected from Cerenkov)  
(3.5 W, 2x300 s)

☹ (very) strong perturbations due to Raman  
from PET material (TEDLAR bag)  
→ Complementary tests required (in open enclosure)

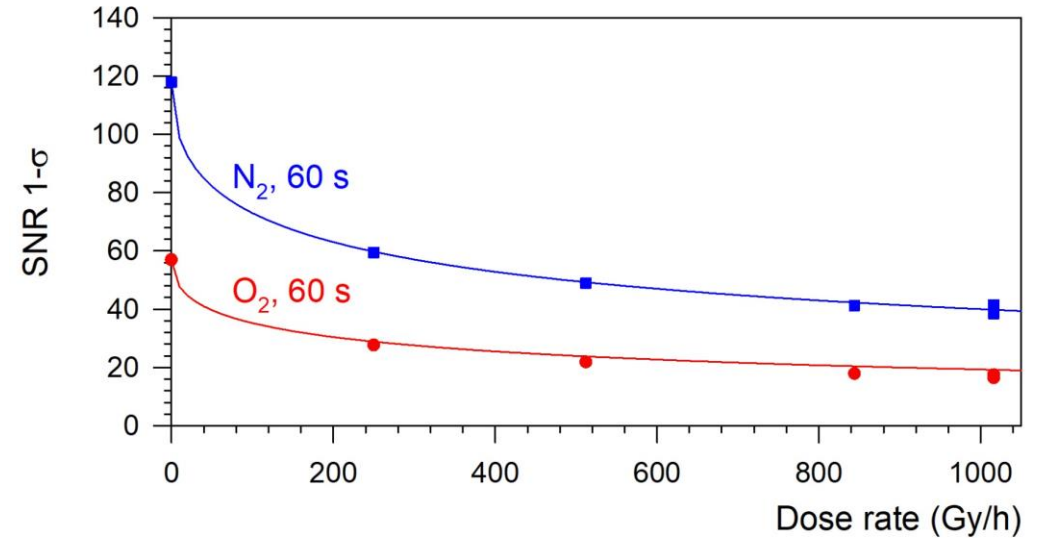


# Radiation qualification (4/4)

Cerenkov-corrected Raman spectra : influence of Cerenkov light on SNR (laser power 3.5 W, 2x60 s)



Evolution of the SNR 1- $\sigma$



$$SNR = \frac{S}{\sqrt{S + 2 \cdot B}} \quad (S = \text{Raman signal}, B = \text{Background})$$

If  $S \ll B \rightarrow SNR \sim \frac{S}{\sqrt{2 \cdot B}} \rightarrow$

$$SNR \approx P \cdot \sqrt{\frac{t}{\dot{D}}}$$

- $P$  = Laser power (W)
- $t$  = Acquisition time (s)
- $\dot{D}$  = Dose rate (Gy/s)

# Limits of Detection (LoDs)

Experimental LODs at SNR = 3  $1-\sigma$  (same time = 600 s = 10 min)  
ICARE (1 W), IRMA (3.5 W)

Dose rate ►	0 kGy/h		0.25 kGy/h	1 kGy/h
Gas ▼	ICARE	IRMA	IRMA	IRMA
	60 x 10 s	300 x 2 s	300 x 2 s	300 x 2 s
H <sub>2</sub>	1.7 %	2 %	3.8 %	6.5 %
O <sub>2</sub>	1.3 %	0.5 %	1 %	1.8 %
N <sub>2</sub>	2.2 %	0.9 %	1.6 %	2.7 %
H <sub>2</sub> O	X	2.5 %	3.4 %	5.7 %
CO	4 à 4.5 %	X	X	X
CO <sub>2</sub>	2 %	X	X	X

*It will be necessary to increase laser power (again) to improve LODs ...*

# Conclusions & perspectives

MITHYGENE Project SUCCESSFUL : PROOF-OF-CONCEPT for remote multigas detection is SA conditions

## Improved nuclearized probe design :

- \_ Sealed enclosure (IP69K) + Brewster window, anti-fogging/anti-aerosol plugged-in module + optical/electrical feedthroughs,
- \_ Local multigas detection (H<sub>2</sub>, O<sub>2</sub>, N<sub>2</sub>, H<sub>2</sub>O, CO and CO<sub>2</sub>) in less than 10 minutes (SAMG),
- \_ Procedure for RIA and Cerenkov correction (*undisclosed / patent pending ...*)

## Qualifications for multigas analysis (H<sub>2</sub>, O<sub>2</sub>, N<sub>2</sub>, H<sub>2</sub>O, CO, CO<sub>2</sub>) [CNRS-ICARE]

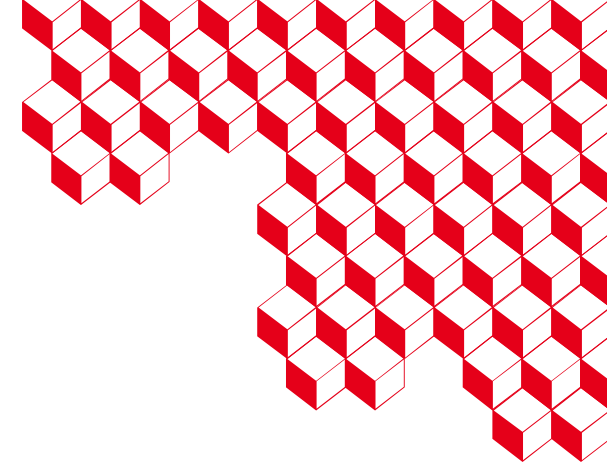
- \_ LoDs are ranging from 1% to 4% (*gas-dependent, SNR = 3 à 1-σ*) for 10-min time and 1-Watt laser power

## Radiation Qualifications [IRSN]

- \_ Validation of the Cerenkov correction algorithm up to 1 kGy/h
- \_ RIA < 6 dB over 20 meters (1.6 MGy)
- \_ Added value : Cerenkov-based real-time distributed dosimetry inside the nuclear containment.

## Perspectives

- \_ Progress in probe design (TRL 6 →), readout unit (TRL 3 →) and DAQ/Algorithm (TRL 3 →)
- \_ Charge-Coupled Device (CCD) will be substituted by photomultipliers (SA-qualification).
- \_ New round of electronuclear qualifications
- \_ Nuclearized Raman probes designed for PWRs and EPRs, also suitable to SMRs and on-board reactors in nuclear submarines.



# Thank you for your attention

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