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Fiber-coupled Raman probe designed for H₂-risk management during a severe accident: Proof-of-Concept for remote gas monitoring in nuclear containments

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During a severe nuclear accident, leading up to core melting, reaction of core constituents with coolant water and Molten Corium-Concrete Interaction (MCCI) both release large amounts of hydrogen (H₂) and carbon monoxide (CO) combustion gases in the containment atmosphere. Depending on local partial pressures of H₂, air (21% O₂, 78% N₂) and water vapor (H₂O), according to Shapiro's ternary diagram, deflagration and detonation may occur with potential deleterious impact over equipments and structures.

To circumvent this H₂-risk, Pressurized Water Reactors of French Nuclear Power Plants (NPPs) are equipped with Passive Autocatalytics Recombiners (PARs). The released hydrogen recombines onto PARs with the oxygen present in the containment atmosphere. PARs only provide partial mitigation because the H₂-generation rate at early times of the accident may exceed the H₂-recombination rate. Two PARs are actually equipped with thermocouples to account for presence of H₂ within the containment building, granted that the O₂-H₂ reaction is exothermic. However, such a temperature measurement does not accurately assess the H₂-risk due to lack of partial pressures data within the containment volume and inability to locate over the Shapiro's diagram.

An improvement in the H₂-risk management strategy therefore requires *in-situ* monitoring of partial pressures of gases of interest at several locations within the containment building in order to account for inhomogeneity and potential local detonation risk. Furthermore, the monitoring of additional gases such as CO and CO₂ provides complementary information about the progression of MCCI. To date, commercial solutions involve overheated gas sampling pipes that transport gas samples out of the concrete barrier into a monitoring module placed outside. The estimation of partial pressures may be strongly biased because of segregation onto the internal pipe surface and it is delayed as well due to gas transport. Finally, the French safety doctrine precludes gas extraction for sake of containment tightness. Therefore, an *in-situ* distributed multigas sensing device is the optimal solution.

Optical-based gas sensing solutions lend themselves to the use of optical fibers for remote monitoring. Unlike absorption techniques (unsuitable for diatomic molecules), all molecules of interest (H₂, O₂, N₂, H₂O, CO, CO₂) exhibit efficient Raman signatures. Spontaneous Raman Scattering conveys many decisive advantages such as chemical-selective, distributed and local gas measurement (mm³). Furthermore, fiber-coupled Raman gas probes may be robust (shock- and vibration-proof) and radiation-hardened.

Within the MITHYGENE Project, CEA LIST, IRSN and ARCYS have designed, assembled and tested a fibercoupled Raman gas prototype probe dedicated to H₂-risk assessment. In view of a proof-of-concept, the Raman probe is fiber-coupled to a single laser (@750 nm, 3.5 W) and a Charge-Coupled Device (CCD)-based imaging spectrometer, both placed away from the radiological perimeter and potentially powered by emergency power supplies in case of power outage (as has happened during the Fukushima's accident). The probe is watertight (IP69K) and equipped with two optical penetration assemblies allowing for laser input and Raman signal output. The optical interface is protected from contaminants and water condensation (Joule heating).

We obtained Raman spectra of air, water vapor, hydrogen, carbon monoxide and carbon dioxide in laboratory conditions. The Limit of Detection (LOD) is typically of several percents for a laser power of 1 W and a response time of 10 minutes. The Radiation-Induced Attenuation is kept below 0.35 dB/m (@800 nm) and 0.2 dB/m (@900 nm) for a total dose of 1.6 MGy, leading to additional losses of 7 dB (@800 nm) and 4 dB (@900 nm) over an exposed fiber length of 20 meters. In accidental conditions, under a dose rate as high as 1 kGy/h, the most detrimental phenomenon appears to be the Cerenkov light stemming from the fiber used for Raman collection. The Cerenkov light superimposes over the useful Raman signal thus degrading both Signal-to-Noise Ratios and LODs. We apply two algorithms to retrieve Raman signals out of Cerenkov perturbations.

The Mithygene Project ends up with a proof-of-concept of Raman probe prototype dedicated to MCCI/H₂-risk management during severe accidents. We estimate the Technological Readiness Level (TRL) of the prototype probe to be about 5 to 6. Complementary data and perspectives will be provided during the conference.

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Abstract (100 words)

Within the MITHYGENE Project, CEA LIST, IRSN and ARCYS have designed a fiber-coupled, watertight and radiation-hardened Raman gas probe dedicated to *in situ* remote monitoring of H₂, O₂, N₂, H₂O, CO and CO₂, in view of MCCI/H₂-risk assessment in NPPs during a severe accident. As a proof-of-concept, the probe is fiber-coupled to a red-emitting laser and a CCD-based imaging spectrometer, both placed away from the radiological perimeter. In accidental conditions, Cerenkov light is the most detrimental phenomenon. We apply two algorithms to retrieve Raman signals. The TRL of the probe prototype is ~ 5 to 6.