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Fiber optics remote monitoring of gas concentrations by Spontaneous Raman Scattering - Application to the H₂-risk management in nuclear containment during a severe nuclear accident

S. Magne¹, S. Nehr¹, X. Buet¹, A. Bentaïb², E. Porcheron³, R. Grosseuvres², E. Studer⁴, R. Scarpa⁴, D. Abdo⁴, J.L. Widloecher⁴, O. Norvez⁴, N. Chaumeix⁵, A. Ruffien-Ciszak⁶, J. Dhote⁶, M. Freyssinier⁶

¹ CEA, LIST, Laboratoire Capteurs Architectures Electroniques, F-91191 Gif-sur-Yvette France

² IRSN, PSN-RES/SAG, BP17, F-92262 Fontenay-aux-Roses, France

³ IRSN, PSN-RES/SCA, BP68, F-91192 Gif-sur-Yvette, France

⁴ CEA/DEN/DANS/DM2S/SFME/LTMF, CEA/SACLAY F-91191 Gif-sur-Yvette, France

⁵ Institut ICARE, 1C Route de la recherche scientifique, F-45071 Orléans, France

⁶ ARCYS, 14 Place Marcel Dassault, F-31700 Blagnac, France

sylvain.magne@cea.fr

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) gas in the containment atmosphere. Depending on local partial pressures of H₂, air (18 % O₂, 81 % 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 (PWR) of French Nuclear Power Plants are equipped with Passive Autocatalytic Recombiners (PARs). Since the O₂-H₂ reaction is exothermic, two PARs are actually equipped with thermocouples to account for presence of H₂ within the containment building. However, such a temperature measurement does not provide any accurate assessment of the H₂-risk.

The H₂-risk management strategy may be significantly improved by performing *in-situ* monitoring of partial pressures of the 3 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 MCCI. Sampling pipes are precluded in order to maintain containment tightness and to avoid gas segregation (measurement delays and deviation with respect to *in situ* partial pressures). Therefore, an *in-situ* distributed multigas sensing device is required for that purpose.

Optical-based gas sensing solutions lend themselves to the use of optical fibers (OF). Among optical-based techniques, the laser absorption technique is not well-suited to symmetric molecules such as N₂ and H₂ because useful optical transitions are dipolar-electric forbidden. Long interaction lengths are thus necessary to compensate for lesser efficiency, in contradiction with the requirement for a localized measurement. Unlike the absorption technique, 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). Raman gas cells are simple, robust (passive optics, no electronics), and may be radiation-hardened. The Raman light is collected by OF and propagates up to an acquisition unit that uses a single laser (@640 nm) and a Charge-Coupled Device (CCD)-based imaging spectrometer (multitrack) leading to cost-effective solution and simplified data analysis. In turn, Raman signals are weak (Raman/laser ~ 10⁻¹⁵) on account of low cross-sections and gas density so that acquisition times are typically of several minutes. Optical interfaces must be protected from contaminants and water condensation (Joule heating).

Within the MITHYGENE Project, CEA LIST, IRSN and ARCYS have designed, assembled and tested proprietary free-space Raman gas cells, fiber-coupled to an acquisition unit placed far away from the radiological perimeter, potentially powered by emergency power supplies in case of power outage (as has happened during the Fukushima's accident). We first describe the calibration procedure performed in climatic chamber (O₂, N₂, H₂O) and in flame-propagation tubes (H₂/N₂) made available by CEA DEN and CNRS-ICARE. Then, we show the results obtained during thermo-hydraulic tests in the MISTRA facility (CEA DEN), representative of a PWR containment at 1/10 scale designed for studying turbulent convective gas flows with condensation (He being often used as gas substitute for H₂). Numerous measurement points inside the containment are simultaneously recorded to set up concentration mapping (temperature, gas composition (Mass Spectrometry), flow rate). Four Raman gas cells were put within MISTRA and the Raman signals were recorded during several runs with the aim of testing their long-term stability. Complementary experiments (temperature, hygrometry, radiation, shock/vibrations) will be described during the conference, providing an assessment of the Raman prototype for H₂-risk management during severe accidents.

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