CONSTRUCTION OF A GAS-MIXING AND ANALYSIS FACILITY FOR FISSION GAS RELEASE AND ANALYSIS STUDIES.

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ABSTRACT

Abstract

The formation of fission gases in the fuel during operation and their rate of release to the grain boundaries and hence to the plenum is a major feature that can limit the linear power rating of the fuel and the duration of a reactor's continuous operation. Much attention has been given to investigating the mechanisms of fission gas accumulation in the fuel matrix and its gradual collection into bubbles, both within the grains and at grain boundaries and how to maximize its retention in the fuel matrix. Considerable work has been done at JRC-ITU and elsewhere looking at the mechanisms and irradiated fuel history effects on fission gas retention/release from the fuel. The current concern for commercial reactors is particularly about fission gas behaviour for MOX or high burn-up UO_2 under operating power transients. Gradual accumulation in the rod free volume would increase the internal rod pressure and may ultimately induce cladding rupture. In extreme cases, such as power transients or accident conditions, sudden fission gas release can result in multiple rods failure and release of fission gas and other volatile fission products to the primary water circuit, forcing reactor shut down.
This paper will look at the construction and preliminary testing at JRC-ITU of a gas-mixing facility and its analytical devices (mass spectrometer, cold traps and gamma spectrometers) along with its connection to a high temperature furnace in view of new campaigns to assess fission gas behaviour during off-normal conditions from commercial or innovative nuclear fuels.

1. Introduction

1.1 Fission Gas Release

The formation of fission gases Xe and Kr in the fuel during operation and their rate of release from rods is a major feature that can limit the fuel's burn-up in operation. Much attention has been given to investigating the mechanisms of fission gas formation and release from fuel pins and their collection in the fuel structure at a microscopic level. Particularly understanding their behaviour and properties is important to fuel fabrication with the best retention properties; at the level of release form the pin, this is particularly a matter of the cladding's performance. In the fuel the Xe, Kr bubbles are created in solid solution and can diffuse to intra-granular or grain boundary bubbles. These bubbles can grow during operation at both sites and at higher temperatures some the large bubbles coarsen (grow) and the expense of the smaller (due to diffusion - Oswald ripening). At the grain boundaries the bubbles grow and link together at the grain boundaries to form tunnels. This provides along with cracking through and between grains the rapid diffusion paths for the fission gas to the outer cladding gap. Here the volatile fission products will remain but the fission gases can accumulate at the free volumes particularly in the plenum of the fuel pin. Here a rupturing of the cladding can result in the release of the fission gases into the coolant. This can be particularly a problem for certain Xe isotopes strongly absorb neutrons (eg. Xe-135, $s=2.6x10^6$ barns) and affect the

reactor's neutron density. It is therefore an important research activity for the safety of fuel and reactor operation.

1.2 Fission Gas measurements

Fission gas release has been assessed by a number of techniques. Heating of samples under a profile that is a simulation of a reactor transient or accident is one technique. Dissolution of a sample of fuel to determine its total content is another approach. Microscopic examination of the fuel either by optical (OM), scanning electron (SEM) or transmission electron microscopy (TEM) can give indications of the fission gas distribution in the fuel at the various magnifications. Scanning Ion Mass Spectroscopy (SIMS) is also able to estimate the fission gas content of bubbles that are opened up during the surface sputtering of the surface by the ion beam. Modelling of the gas content can also provide an estimate that can enable the relative size of the release to be assessed.

However for measurement of fission gas releases on the macro scale, it is common to take a small piece of fuel probably still cladded to minimise fuel damage and to subject it to a transient heating expected in operation. Here the interest is to find the radial distribution of the fission gas in the fuel and the distribution between gain boundary and intragranular bubbles. The OP or SEM examination can assist by showing if the the intergranular bubbles have linked together and enabled open pathways for the intergranular fission gas through the gain boundaries to the cracks for collection at the outer gap or plenum. At higher burn-ups when the cladding has undergone creep on to the fuel the gap is no longer open and easily connected to the plenum. This means that fission gas estimates from pin puncturing techniques can be less reliable as complete extraction of the plenum and gap gas content becomes increasingly difficult. The use of microprobe analysis of fuel sections can detect the Xe fission gas in solid solution and the X-ray fluorescence spectroscopy can detect both Xe in solid solution and Xe in bubbles. This also enables the Xe distribution in the solid and in the bubbles to be determined along the radial axis ie the temperature profile.

2. Fission Gas Facility

ITU has undertaken a considerable new construction of a fission gas measurement facility, in order to match oven facilities installed in the hot cells. The first purchase was a gas-mixing facility, This was then followed by a purchase of cold-traps with N_2 flasks with electronic temperature monitoring and automatic refilling of the cold traps from a central large (120l) Dewar vessel. The cold traps were then built into a lead shielded cabinet along with gamma spectrometers able to measure the activity captured in the cold traps.

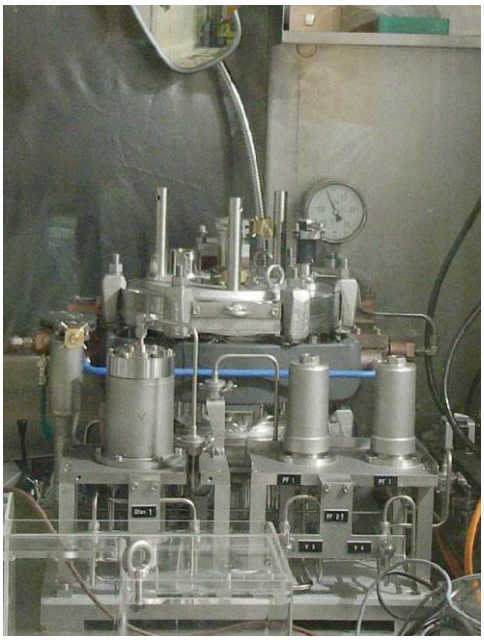

Fig. 1 High temperature furnace in the hot cell during installation in the hot cell

Finally it was also decided to add a mass spectrometer linked to the gas–mixing lines so that further analysis of the fission gases could be made. The whole facility was placed in the basement because of space limitations in the laboratory. However there are also working restrictions in the basement that would often cause sizeable delays in the construction.

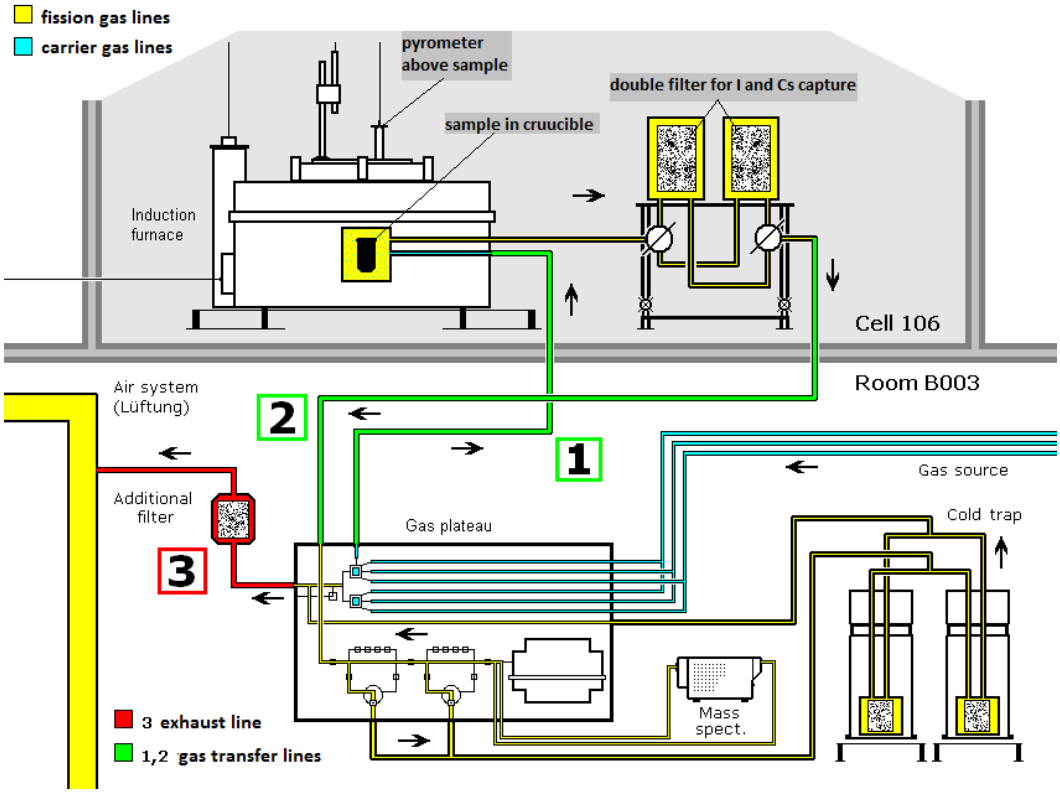

Fig. 2 Complete ITU facility for fission gas release measurements showing the gas circulation between the high temperature oven in the hot cell and the gas-mixing station attached to N_2-cooled cold traps with gamma spectrometers and mass spectrometer in the basement.

The facility required 3 computers with the appropriate software to monitor and conduct experiments along with a final computer above in the laboratory to be able to follow the experiment remotely. The computer controlling the furnace is also in the laboratory. The high temperature furnace in the hot cell is shown in Fig. 1 and the scheme of the complete facility is shown in Fig. 2.

2.1 Gas mixing Facility

The gas-mixing facility is a specially made for ITU by SEMPA systems, Dresden following ITU specifications to be capable of mixing the carrier gases to a desired composition by control of the relative flow rates before entering a high temperature oven located in a hot cell. The gas line should pass over the samples in a crucible entraining any released gases from the sample and return after passing over a water spiral to remove volatile species such as Cs and a second filter of activated charcoal to remove iodine and other volatile fission products, before passing over an absolute filter. The carrier gas with released fission gas traces should then return to the facility to be passed over a liquid N_2 (boiling point -196°C) cold trap with a activated charcoal cold trap to freeze out the Ke and Xe fission gases (solidification point -157°C and -112°C resp.). The carrier gas is then returned to the facility for passing into the active gas exhaust line. A schematic outline of the system taken from the controller software included in the system is shown in Fig. 3.

Another function of the gas-mixing station is to capture the fission gases. There also specific circuits for calibration; in this circuit the carrier gases can then sweep out the released calibration gas from a registered source for passing over and collecting in the cold trap for calibration. There is also the function to allow the cold trap contents to be reheated and released into a closed circuit of known volume where the gases can be distributed evenly. Then the well-mixed fission gases can then be sampled and analysed by mass spectroscopy.

The final function was to connect with the exhaust system of the hot cells and to use carrier gases to purge the fission gases into the hot cell exhaust line for dilution and release. All systems have been preprogramed into the system and require a single command to switch all the necessary valves to create the correct circuit required for specified function (eg. measurement or calibration)

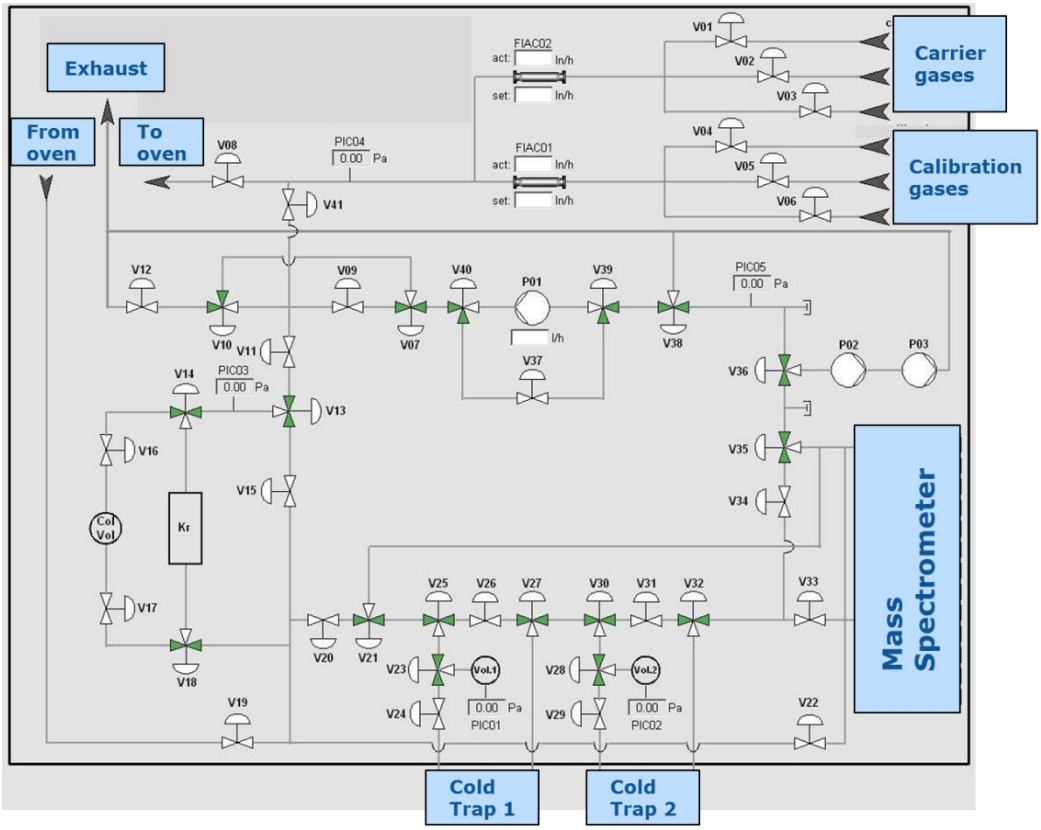

Fig 3 Flow scheme for the Gas-mixing facility as shown on the Software for mixing system monitoring

2.2 Cold Trap Cabinet

The two liquid N_2 cold traps are double steel–walled vessels with connections to evacuate the inter-wall volume and so be able to maintain extreme temperature differences. This was designed to maintain liquid N_2 temperatures for a minimum of a week. A double cold trap system was constructed by KTG, Karlsruhe, and delivered with an electronic liquid N_2 monitoring system to activate valves for refilling the cold traps from a connected 120l Dewar vessel. The double cold trap system (the second trap to ensure capture of all the fission gas released) also needed mounting in a frame where the gamma spectrometers were correctly positioned to optimally measure the accumulated activity of the Kr-85 content in the fission gases. This required lead shielding to enable protect personnel and electronics against the accumulated fission gas activity and any stray background in the basement. Fig. 4 shows the lead shielded cabinet opened to reveal the double-walled vacuum cold traps inside. In Fig. 5a) and b) details of the charcoal cold traps to capture the fission gas are shown inside the specially constructed double-walled vacuum flask to contain liquid nitrogen and keep the charcoal trap in the gas lines chilled to ~ -197°C, while allowing the close positioning of the gamma detectors for good signal sensitivity.

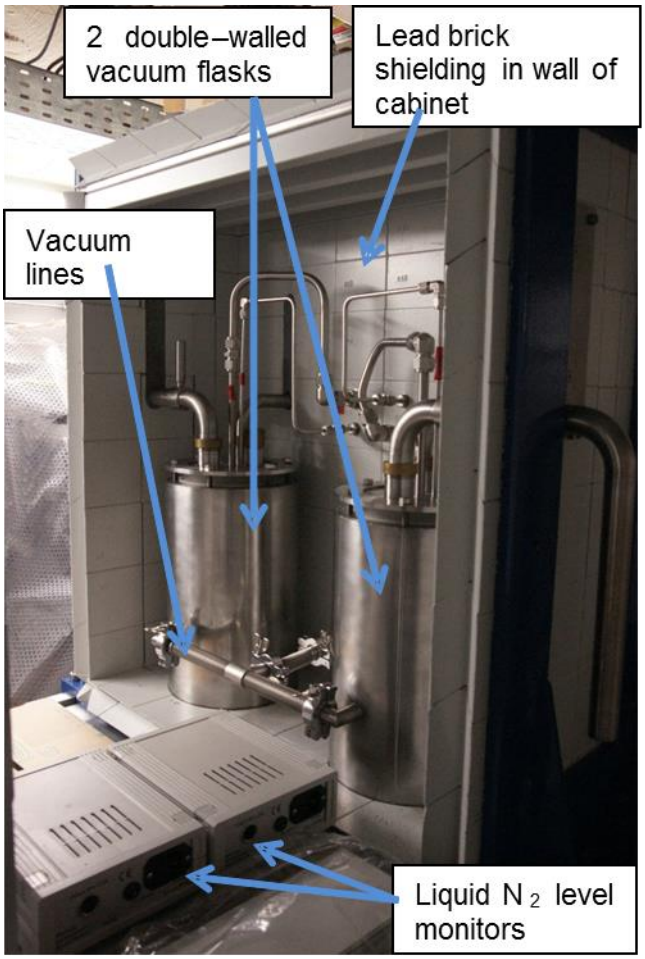

Fig. 4 photograph of the lead–lined cabinet showing the cold trap inside the lead shielding and the 2 liquid nitrogen level monitors at the front.

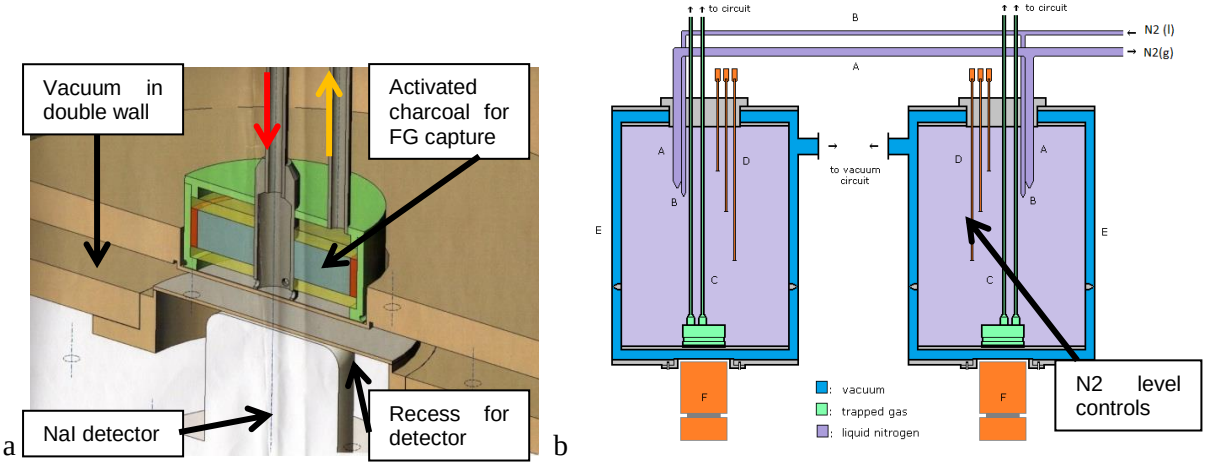

Fig. 5 a) detail of the KTG Dewar vessel with its double wall and the cold trap filter with an activated charcoal for collection of the fission gases; b) diagram of overall layout of the cold traps, their vacuum and the circulation of the liquid N_2 cooling system and its level controls. This ensures efficient freezing out of the fission gases on the charcoal filter for detection of the Kr-85 gamma line by NaI detectors

2.3 Gamma Spectrometers

The next feature is the gamma spectrometers. NaI has been the most commonly used as a relatively unspecific detector but with high sensitivity. 2 Ortec Bicron NaI (5cm dia. crystal) detectors have been purchased along with the Maestro software for spectrum collection and

evaluation. If it is not certain that the gas composition will contain only Kr-85, then it may be useful to have a more accurate detector (even with lower sensitivity) such as high purity Li with Ge (HP Li-Ge) detector or an intermediate La Br detector. We intend to purchase HP Li-Ge crystals in the future. The detectors are linked to an independent PC (not linked to any other system PC) to set-up perform and log measurements as well as performing spectral analyses.

2.4 Gas Mass Spectrometry

A Pfeiffer GSD320 mass spectrometer is connected to this circuit for the purpose of analysing a small fraction of a well –mixed gas circulating in a closed circuit. This is able to perform relative analyses of the gas composition and accurate trace contents analyses down to 1 ppm. The device is also linked to a PC operating the Pfeiffer commercial software to set-up, perform measurements and store results.

The determination of the relative ratios of the Xe isotopes and the Kr isotopes that make up most of the fission gases observed in the irradiated fuel releases is particularly important when combined with the absolute determination of the Kr-85 by gamma spectroscopy of the fission gases in the cold trap. The combination of the information enables all Xe and Kr isotopes to be calculated quantitatively. This again together with data from puncturing gives further information on the performance of the fuel at this burn-up in comparison with the puncturing gives the total releases to date. However it does not indicate at which stage the releases have occurred or if it could be related to any particular feature of reactor operation.

2.5 Controlling Systems

The mass spectrometer sits on a table beside the shielded cold trap cabinet alongside its controlling PC and the PC for the Gamma detectors. The 3rd PC controlling the gas-mixing facility is also placed on this table. As noted above the γ spectroscopy PC has to be independent (programme operation requires administrator rights) and is not connected to any system. The gas-mixing and the mass spectrometer computers are linked to a final computer upstairs adjacent to the laboratory, so that it is possible to perform an experiment with the oven and be able to check there from the laboratory the condition of the measurement systems especially the carrier gas flow rates over the sample in the hot cell furnace and the MS results. See Fig. 6

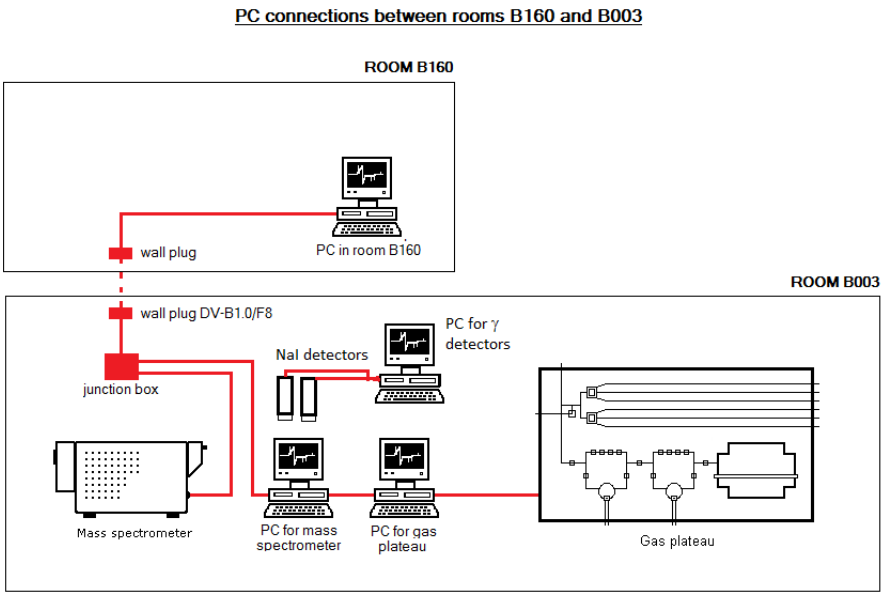

Fig. 6 showing the connections between the computers in the fission gas measuring system.

3. Latest developments

During the setup of the cold-trap Dewar vessel and mass spectrometer with the 2 other computers, the net dose rates were checked in the vicinity of the gas–mixing table. It was found that there was a ventilation pipe (and also filter housing), locally increasing doses to levels over the acceptable limit: thus 0.38mSv.h^{-1} were noted on contact of the overhead piping. Therefore it was necessary to put up a shielding for the piping to reduce the dose rates. The re-measured dose levels were approximately one-third to one-fifth of the initial levels (see fig. 7). This necessary monitoring of the background activity levels has naturally added considerable work and slowed down progress in the construction and co-ordination of the whole facility.

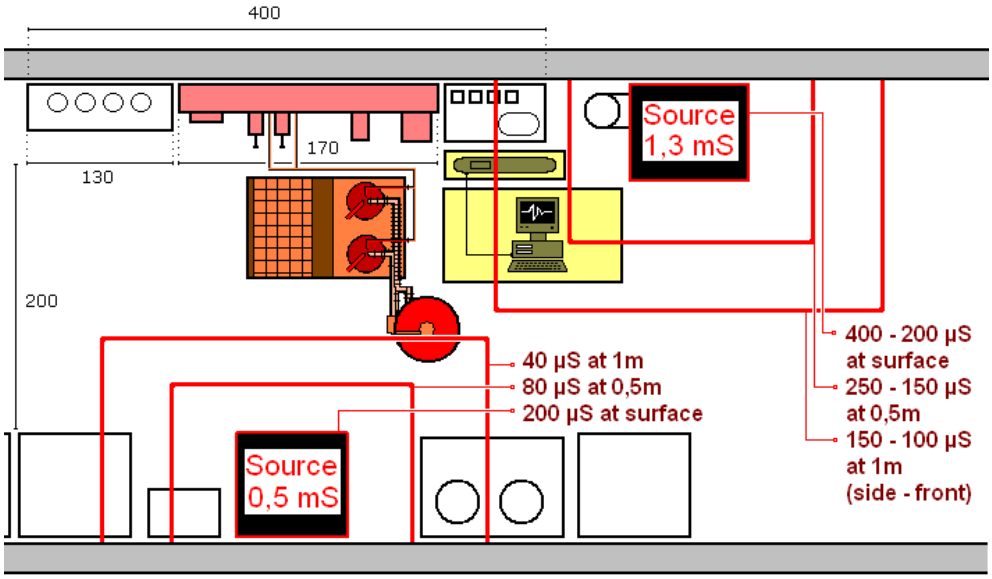

Fig. 7 Diagram of the lowered dose rates in the basement below the Hot Cells at points near the gas-mixing tableau, the cold traps and the PC desk of the FG facility.

In addition it was also decided to displace the PC desk 5m away so as to reduce the activity further. The current arrangement of this equipment is shown in Fig. 8.

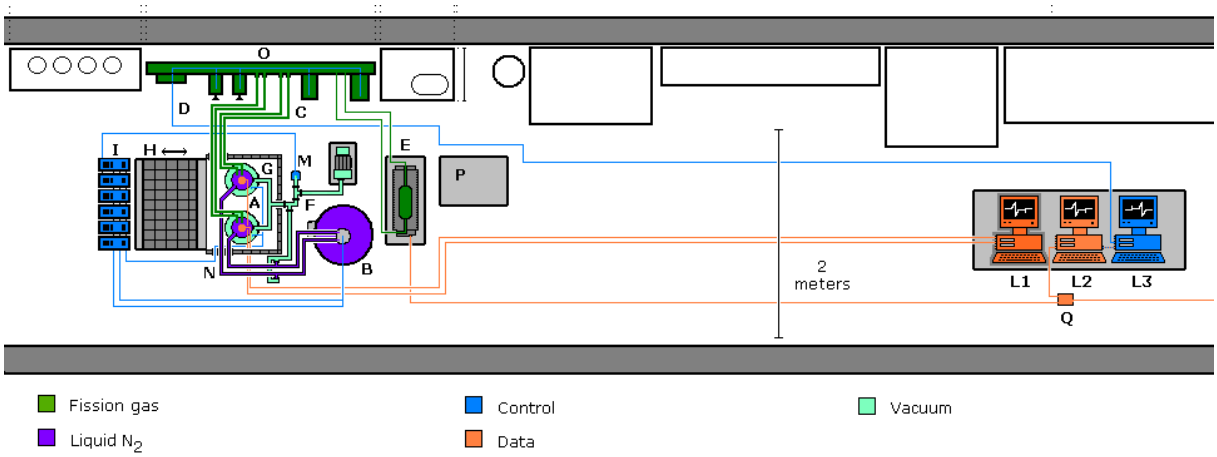

Fig. 8 Display of the current arrangement of equipment in the room below the Hot Cells, with the PC table moved to a corner of low activity. A: NaI γ-detectors; B: Liquid N_2 container (150L); C: gas lines between cold traps and gas plateau; D: PC/Tableau/ MS gas & data links; E: mass spectrometer; F: Vacuum Pump; G: shielded Dewar containers of cold traps; H: mobile lead shields; I: liquid N_2 level control units; L: control PC's (L1 not connected to net); M: Pirani vacuum gauge; N: vacuum-insulated N_2 feed tubes; O: gas distribution plateau; P: table; Q: connections to local net.

Recently the manufacturers of the gas –mixing facility have been able to come and verify the operational fitness of the equipment and of the software. This has resulted in replacing the computer and connection cable with the gas-mixing facility and a reorganisation of the exhaust line. So a commissioning trial of the gas-mixing facility and other parts of the fission–gas facility will soon be possible. The oven has been tested in isolation as have the gamma detectors and the mass spectrometer and the liquid N_2 monitors. The final testing will be to use gas-mixing, gamma detection and mass spectrometry function together and to check the interfaces function smoothly. Finally the links between the computers will be checked, and specifically that the gas facilities in the basement can be adequately monitored in the laboratory above while a furnace test is underway in the hot cell.

4. Conclusions

ITU has been steadily constructing a new fission gas research facility in the Hot Cells. This has required a new gas–mixing facility, new cold traps with liquid N_2 control system and attached gamma spectrometers and a mass spectrometer in the basement for linking up with a high temperature furnace in the Hot Cells above. The construction has taken much time and the requirement to monitor activity levels in the basement has inevitably slowed progress. This has also resulted in an additional requirement to optimise the position of the PC's in order to locally monitor the operation of each component. After the commissioning of each component, all components in the basement room will be tested, before passing on to a combined 'cold' test using the furnace and the basement components. If all previous tests are successful, then one could pass on to a first active 'commissioning' test.

5. References

[1] E. Fontana, P. D. W. Bottomley, V.V. Rondinella, Experimental setup for the release and study of fission gas, , 49th Annual Meeting 'Hot Laboratories & Remote Handling Working Group' : HOTLAB 2012, 24-27th September, CEA Marcoule, France.