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DIRECTION DES REACTEURS NUCLEAIRES
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Partition of grain boundary and matrix gas inventories in nuclear fuel: the ADAGIO facility

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THE ATOM, FROM RESEARCH TO INDUSTRY

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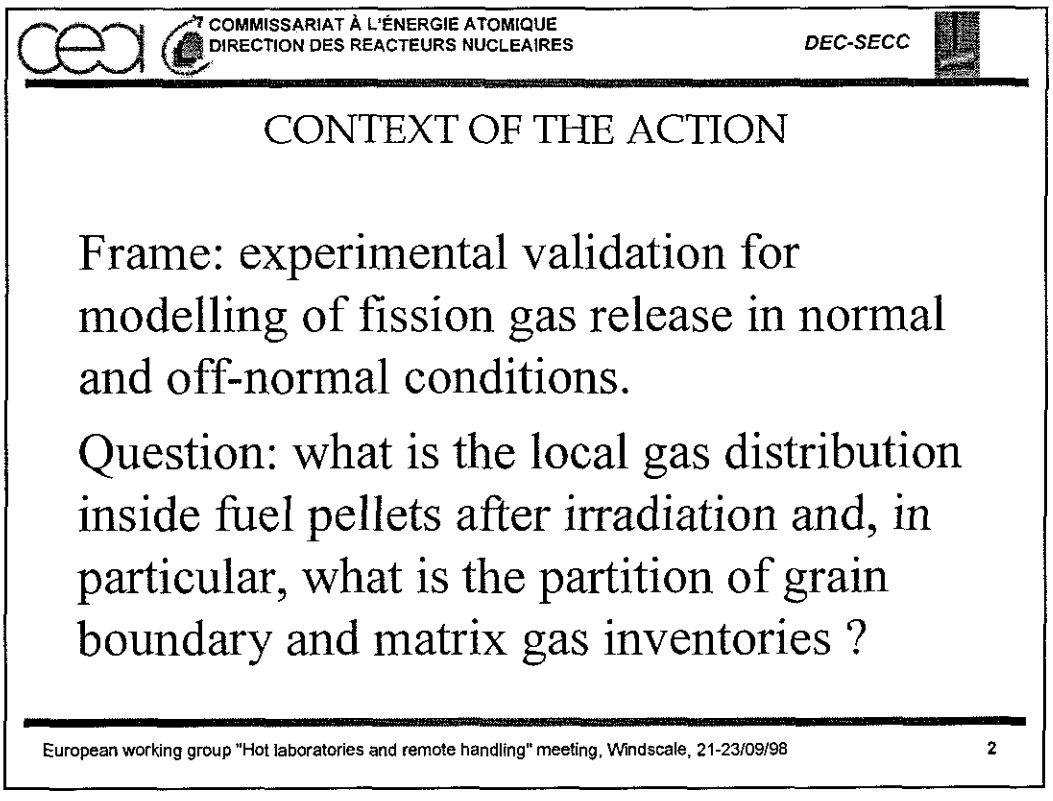
ABSTRACT

The improvement of fission gas release modelling in nuclear fuel performance codes needs experimental validation regarding the local gas distribution inside fuel pellets. This information is important in both normal and off-normal conditions.

In this frame, an experimental programme has been initiated by CEA, EDF and FRAMATOME Nuclear Fuels using irradiated PWR fuels. The process is closed to the method used in AECL Laboratories on Candu fuel. A pre-test reirradiation is performed in an experimental reactor in order to rebuild a radioactive tracer of matrix fission gas (i.e. ^{133}Xe) in low temperature conditions preventing any diffusion process. The first step of the experiment is core-sampling by ultrasonic drilling that allows a well-known location of the samples (central or peripheral zones). The next step is an oxidation process under controled temperature and air atmosphere. At low temperature (450°C) the transformation of UO_2 into U_3O_8 takes place and leads to the opening of grain boundaries. This process leads to the total release of ^{85}Kr at grain boundaries. The part coming from the matrix is evaluated thanks to the tracer. Increasing temperature induces a complete release of both intergranular and dissolved gas.

To perform these tests, a new facility has been built in the LAMA Hot Laboratory, in the Grenoble CEA Centre. An inductive furnace able to reach 1700°C, a specific on-line gamma measurement unit and cryogenic gas trapping devices are the main elements of the experimental facility. The post test characterization includes gamma measurements and gas chromatography of the collected gas, and scanning electron microscopy of the powdered fuel.

In the early 1998, several tests have been performed up to 1450°C on an high burn-up fuel submitted to a power transient. Results clearly indicates a larger grain boundary inventory in the central part compared to the periphery. An extension of the program is planned with MOX fuel.



The ADAGIO facility is implemented in the LAMA hot laboratory of the CEA Grenoble.

The word ADAGIO is a French acronym for Discriminating Analysis of Accumulation of Intergranular and Occluded Gas.

This presentation shows principles, techniques and devices of this experiment which is working since January 98.

Fission gas release must be predicted by codes. These predictions have to be realistic for a maximum of different kinds of fuels and working conditions. But, the data base is limited especially under transient conditions.

The experimental programme has two main objectives: firstly, to extend the data base in the range of high burn-up and transient conditions and, secondly to develop more accurate mechanistic models.

In particular, for ADAGIO, the purpose is to quantify the partition of grain boundary and matrix gas inventories.

The first tests completed at the beginning of this year had to demonstrate the feasibility of the experiment.

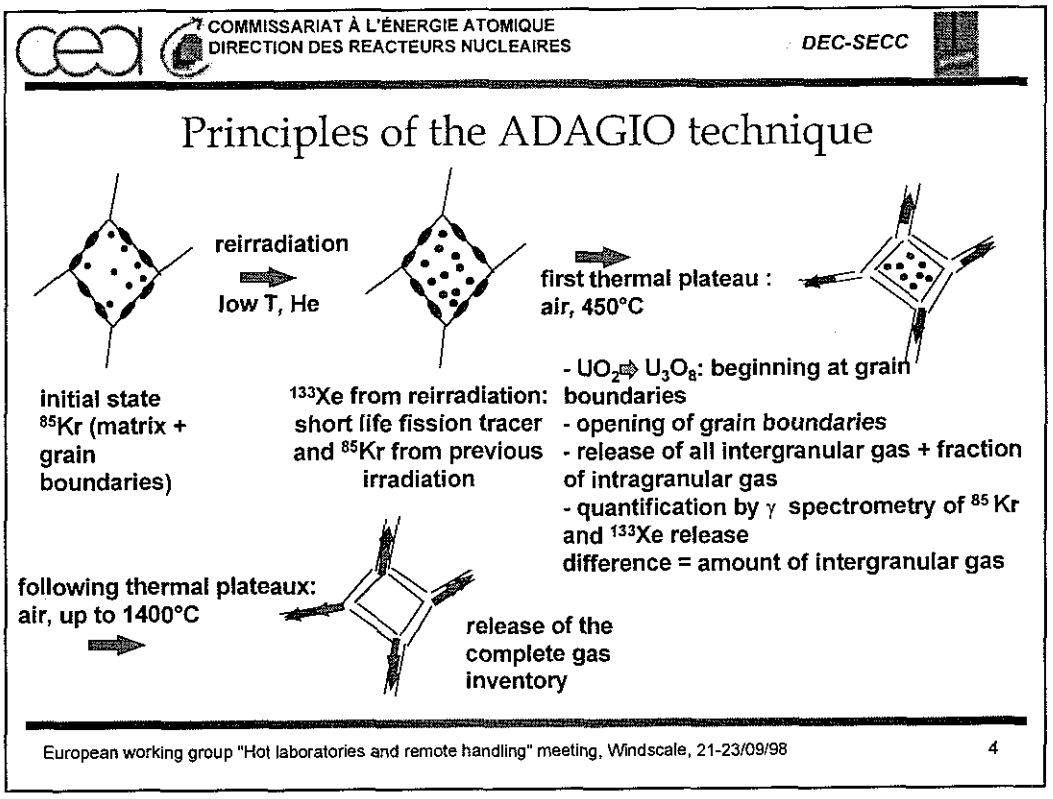
The origin of the ADAGIO technique

A method developed at the Chalk River Laboratory (AECL Research) for Candu fuel.

This method has been adapted for our requirements using our experience of fission gas measurement.

Principles used for the ADAGIO experiment have been previously perfected at the Chalk River Nuclear laboratory which is working for the Atomic Energy of Canada.

The ADAGIO experimental device has been adapted from the Canadian experiment using our own materials and experience of radioactive gas measurement.



Samples studied are small parts of a pellet of nuclear fuel irradiated in a nuclear plant at a BU near 50GWd/t.

At the grain level (about 10 µm), and after several cooling months, the only remaining radioactive fission gas is ^{85}Kr. This gas is localised for a part in the matrix and for the other part at the grain boundaries. This is the initial state.

For the experiment the pellets have to be irradiated again in order to create short life fission gas. The irradiation is done at low temperature and in an inert atmosphere in order to minimise the diffusion of the gas and not to oxidise the fuel respectively. For our tests, pellets have been irradiated in an experimental reactor at 10W/cm during a week, the temperature was less than 350°C. The gas created is principally localised into the matrix because it does not have enough time to diffuse to the grain boundaries. The most interesting isotope is ^{133}Xe because it has a not too short half time (5.25 d) and it is well detected by gamma spectrometry.

After the sampling stage, the heat treatment can be done. The first plateau at 450°C under air flow, induces the oxidation of UO_2 into U_3O_8. This phenomenon starts at grain boundaries. U_3O_8 is less dense than UO_2, then grain boundaries are cracked and intergranular gases are released. This release is followed by on-line gamma spectrometry. The difference between ^{85}Kr an ^{133}Xe gives the amount of inter-granular gas. The first thermal plateau is maintained till the total oxidation of the fuel. The sample is became a very fine powder of U_3O_8 grains.

The following thermal levels realised up to 1400°C produce the release of complete gas inventory by diffusion.

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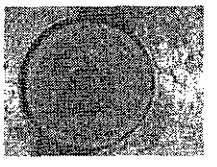
The core sampling drilling of the pellet

- Partition of inter and intragranular gas inventories is different inside the pellet
  - ➠neccesity of sample precisely located
  - ➠ultrasonic core sampling drilling :
    - pellet not coated
    - thickness of the tool = 0.3 mm
    - diameter of sample = 3 to 7.1 mm
    - hight of the sample = 4 to 5 mm



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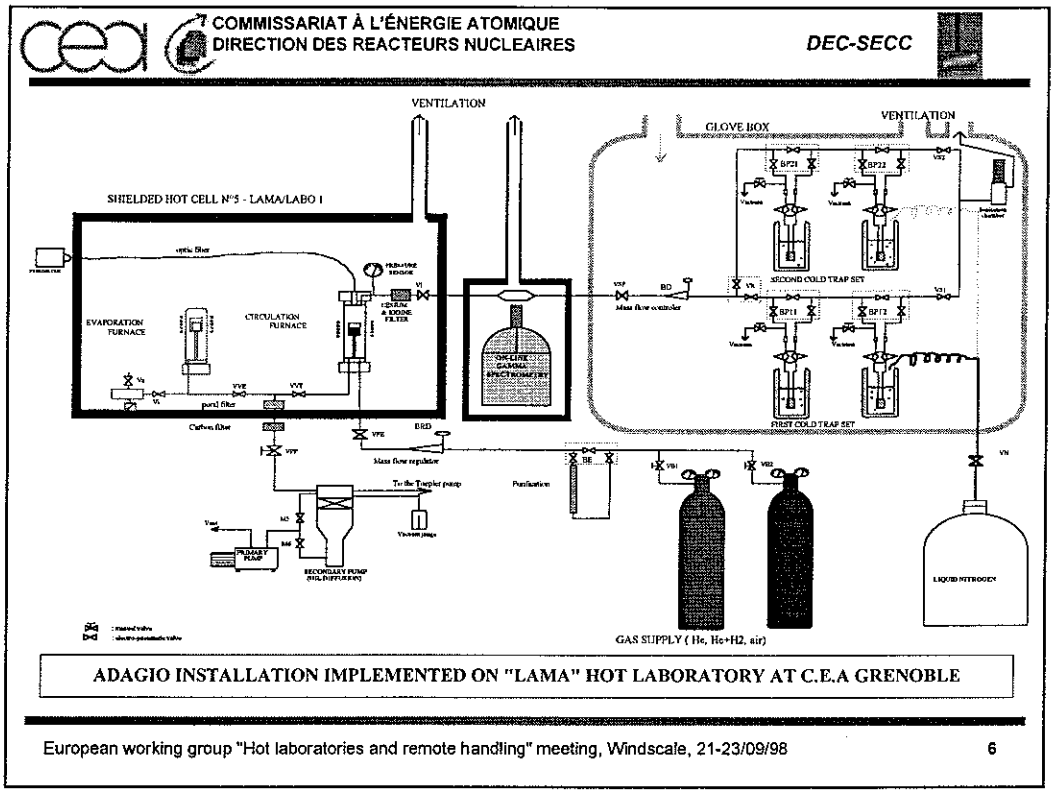
We have already described the reirradiation and the heat treatment but we did not talk a lot about the sampling which is very important.

One of the major interest of the experiment is to analyse samples precisely localised in the pellet because the partition of inter and intragranular gas is different.

The sampling is performed by ultrasonic drilling. The thickness of the tool is 0.3 mm and the internal diameter of the hole is between 3 to 7.1 mm. The hight of the sample is 4 to 5 mm. This operation is done on a non coated pellet because coating could held up the fuel oxidation.



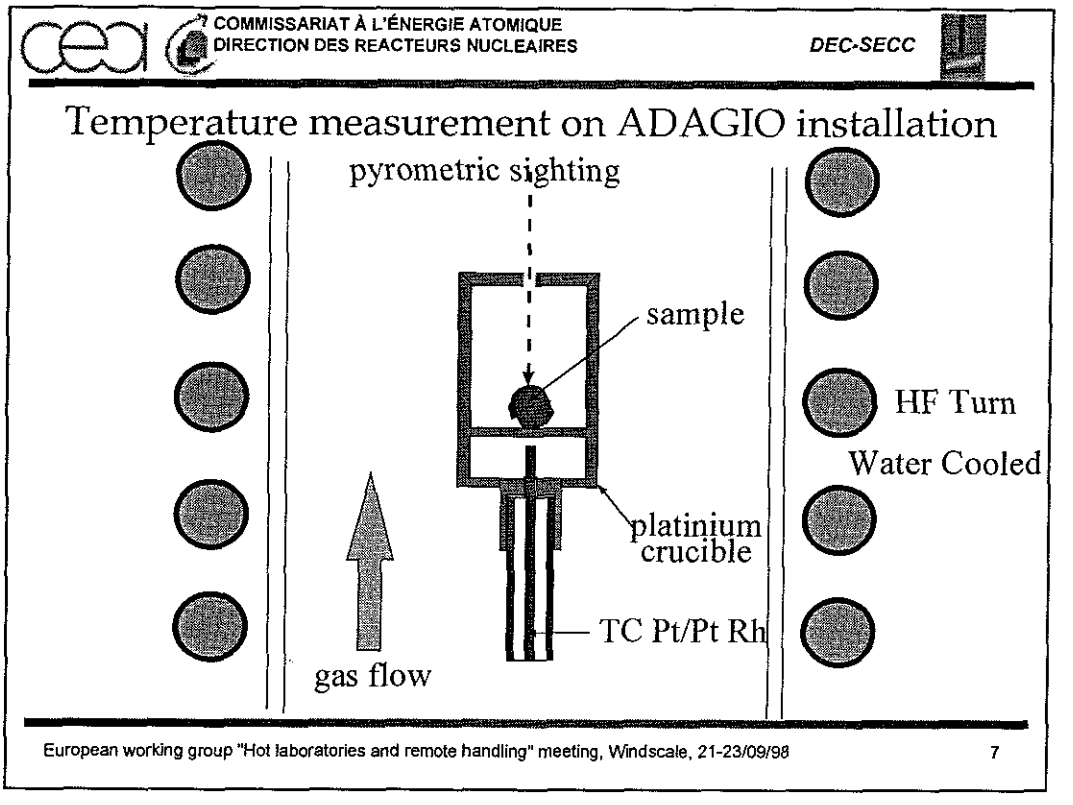
This schema describes all the equipments of the heat treatment device with on-line and cumulative gas release measurements. It could be divided in three main parts: the shielded hot cell, the gamma spectrometry monitoring and the glove box.

The dry air provided by cylinders is regulated at the entrance of the hot cell. The high frequency furnace is constituted by water cooled copper turns where 100 kHz electric current is circulating. An inducing current takes place is the metallic crucible and warms it. A quartz tube provides airtightness of the furnace. The sample is placed into the crucible. Temperature, pressure and flow are recorded.

After the furnace, air loaded with radioactive tracers passes through filters. These filters constituted by reticulate carbon and fine poralinox stop emitted aerosols in particular iodine, caesium and ruthenium.

After filters, radioactive rare gases pass in front of the gamma detector where the gas release is measured.

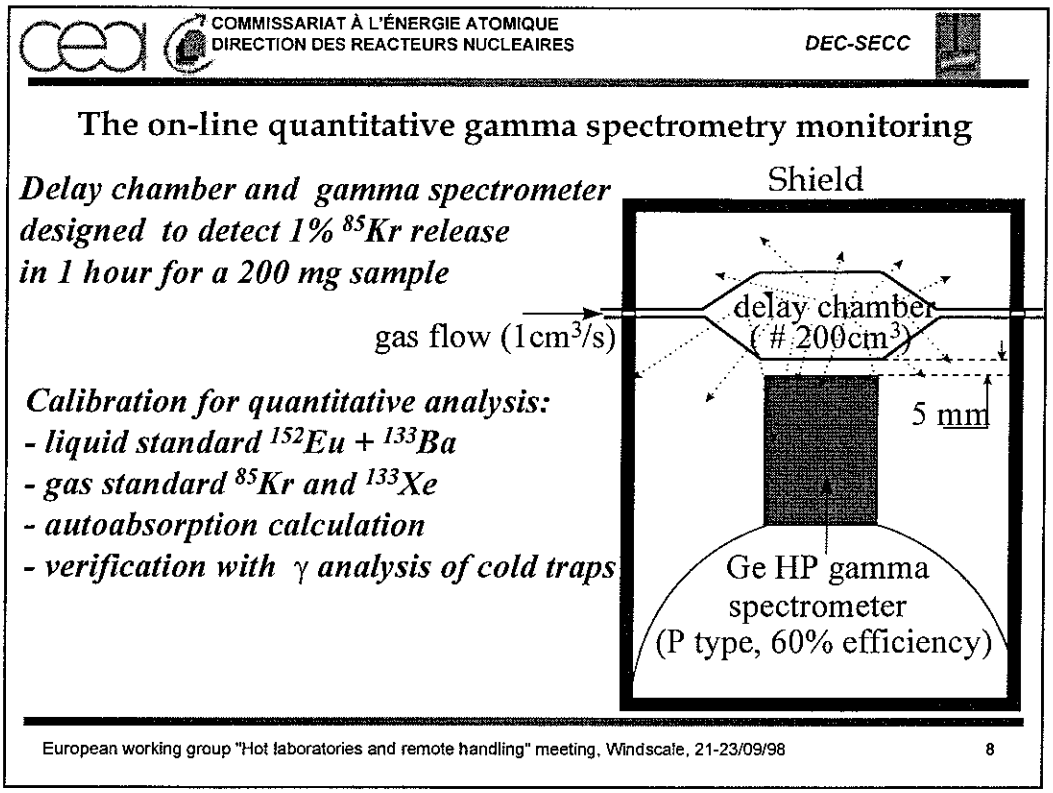
Then the gas is coming into the glove box. Inside, there are two cold trap sets where xenon and krypton are trapped. During experiment one set of trap is working and the second one is prepared in case of emergency. The purified gas is controlled at the exit by an ionisation chamber.



Temperature measurement is performed by two ways: pyrometer and thermocouple.

The thermocouple is placed into a small chamber of the crucible. This chamber is supposed to be at the same temperature as the chamber of the sample.

A two wavelength pyrometer gives another measurement by direct sighting into the chamber of the sample. It works only above 1000°C.

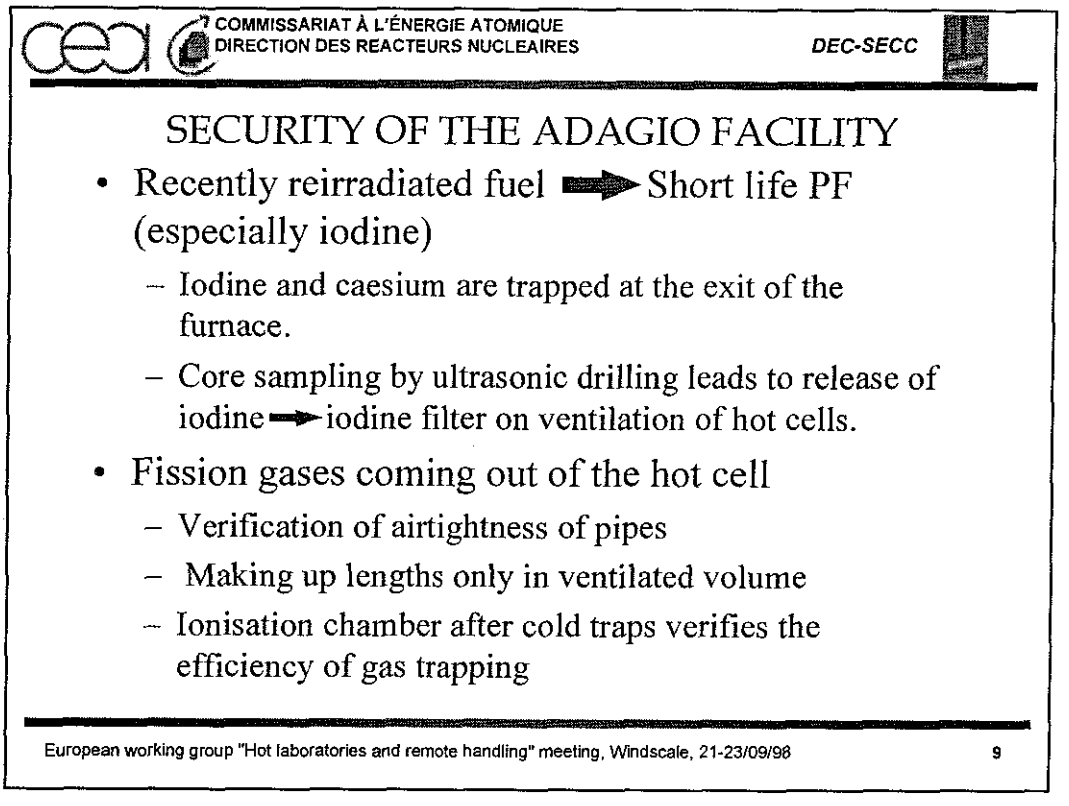


This point is the most important of the ADAGIO installation.

This equipment has been designed to be able to quantify 1% of ^{85}Kr release in 1hour for a 200mg sample with a BU of 50 GWd/t. In this case, that means an emission of only 8 gamma rays per second and per cm^3 of gas. So, a very sensitive detector has been chosen and, in order to have a good signal to noise ratio, the detector was shielded. In the same spirit, the geometry and the position of the delay chamber have been studied. The geometry minimises dead volume and it is placed very close to the detector. The wall thickness of the capacity is 0.5 mm in front of the detector.

Calibration for quantitative analysis of this equipment is a difficult operation. We fill the capacity with different liquid and gas standard to have a realistic calibration. Measurements on liquid standard are corrected by autoabsorption calculation.

Typically, one spectrum is recorded every 3 minutes. This frequency is adapted with the counting rate to have the best possible sampling of the release signal. A deconvolution calculation has been also performed. It permits to obtain the release function of time at the sample level from the release measured at the delay chamber level.



The analysis of recently irradiated fuel induces some security problems.

The essential point is the presence of radioactive iodine into the sample. This nuclide is very volatile, so drilling and heat treatment generate contamination. For drilling, the hot cell have to be equipped of iodine filters on ventilation. For heat treatment, filters are placed at the furnace exit. These filters have to trap caesium and iodine but not fission gases. We have performed several tests to optimise this point.

During the experiment, fission gases are coming out of the hot cell to go to the glove box. A part of the pipe passes in the laboratory. In this part there are not making up lengths and the airtightness of the pipe is verified before experiment. For radioprotection, a radioactive gas detection is implanted in the working zone.

The ionisation chamber placed after cold traps is very useful to be sure that cold traps are working correctly. The current measure gives only an indication and cannot be use for quantitative measurement.

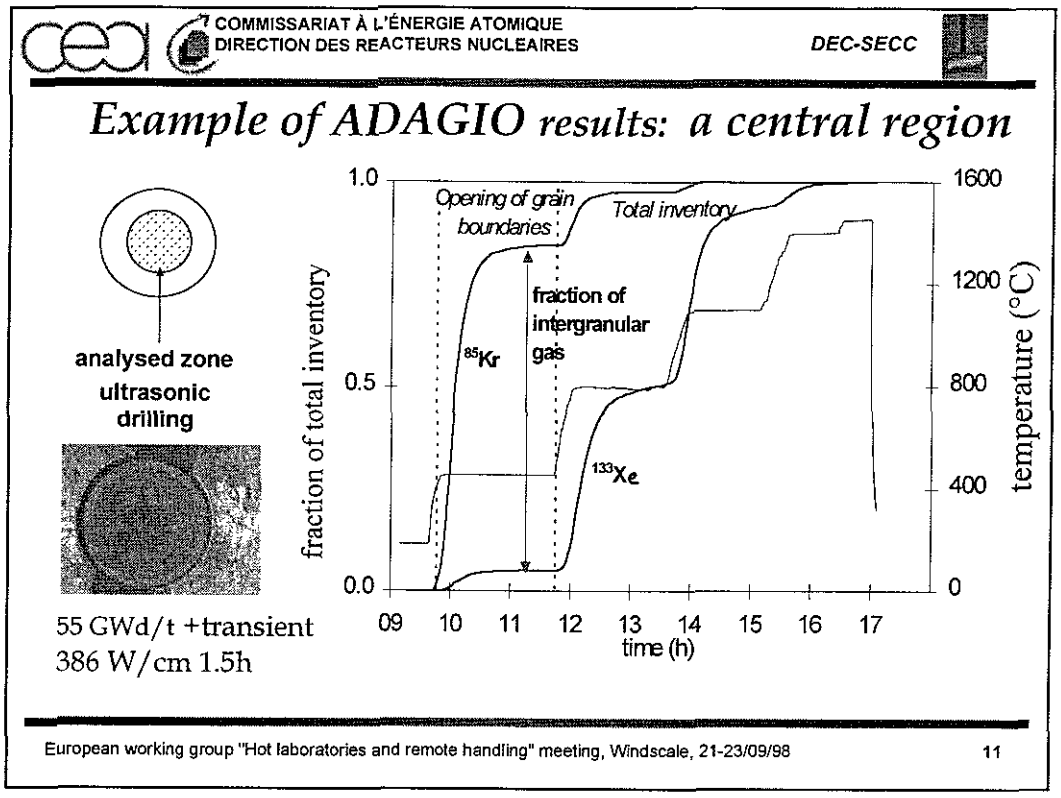
THE PROBLEM OF TRAPPING

- PROBLEM: trapping of krypton ($T_{liquefaction}$ = -152°C) and xenon ($T_{liquefaction}$ = -107°C) with charcoal on air circulation without stopping oxygen ($T_{liquefaction}$ = -183°C) => liquid nitrogen is too cold (-196°C).

preparation of carbonic snow cooled at around -150°C with liquid nitrogen

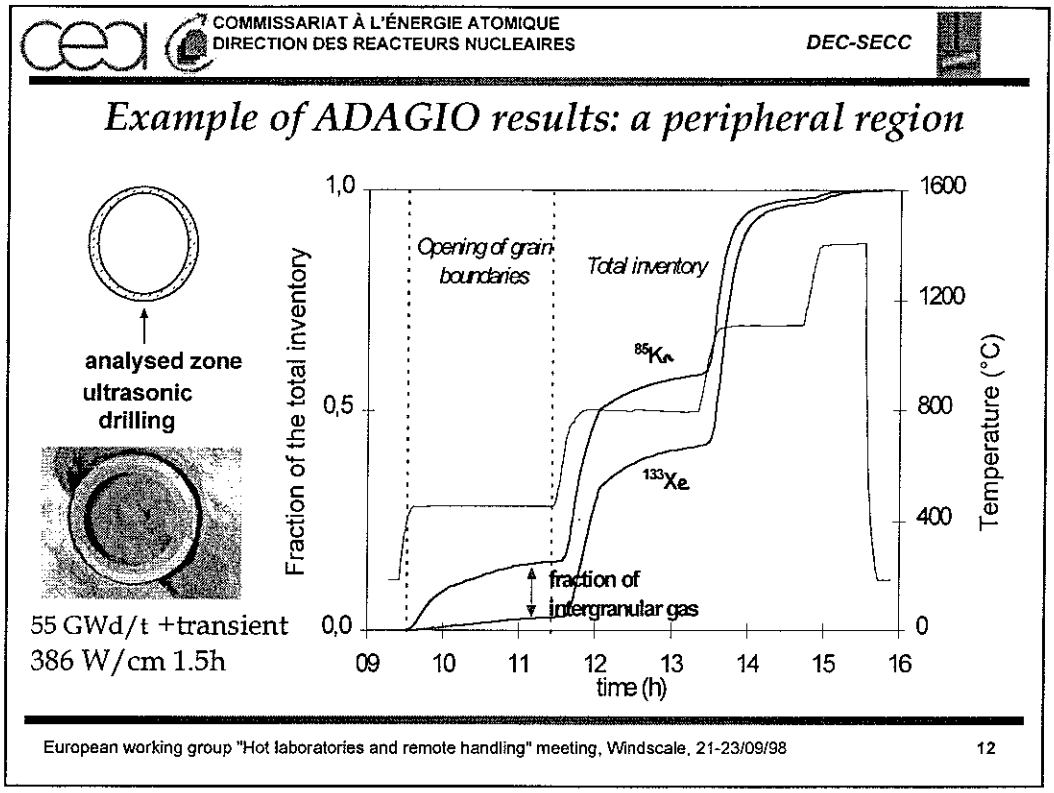
The laboratory has a long experience of fission gas trapping in charcoal or molecular sieves cooled with liquid nitrogen.

But ADAGIO is a special case because it works under air circulation and we usually operates with helium. The problem is that oxygen is trapped at liquid nitrogen temperature and this large amount of oxygen into the trap (about 700 cm^3 per hour) is not acceptable when the trap is reheated at ambient temperature. During the first tests of the installation we tried to trap at carbonic snow temperature (-78.5°C). We observed that xenon was trapped but krypton passed through the charcoal; it was only slowed. So, we made a colder preparation of carbonic snow cooled with liquid nitrogen at around
-150°C. This preparation gives better results in trapping krypton but, when the experience is quite long (several hours) a non negligible part of krypton is carried out of the traps.



In the two examples, the analysed pellets have a burn up of 55 GWd/t and have been submitted to a transient power of 386 W/cm during 1h30.

In this transparency, you can see the location of the sample and the release of ^{85}Kr and ^{133}Xe during heat treatment. The fuel is sampling in the central region of the pellet (hole diameter is 4 mm). At 450°C about 80% of krypton and only 5% of ^{133}Xe are released. The difference is the fraction of intergranular gas. The total inventory is obtained with heat treatment up to 1400°C.



Here, you can see another result of a sample taken in the peripheral zone of the same pellet.

The release curves show that the fraction of intergranular gas is smaller than the fraction observed on the central zone sample. This result is logical because, during irradiation, the central zone is warmer than the peripheral zone and fission gases can migrate and precipitate at grain boundaries.

CONCLUSION

- quantification of the partition of grain boundary and matrix gas inventories in localised samples completed for six tests under air
- ways of progress :
  - better efficiency for krypton trapping in air circulation
  - more precise location of sample

The first set of tests performed in January and February 98 have reached the objective which is to show the feasibility of the experiment. The quantification of gas release and the partition of grain boundary and matrix gas inventories have been done on different samples.

But two principal ways of progress are identified:

- the trapping of krypton have to be improved because gamma analysis of the traps must give the cumulative release in order to be compared to the on-line measurement.

- core sampling drilling on irradiated sample is not perfectly controlled. Sometimes the cylindrical sample is not well centred in the pellet. This point will be improved with the installation of a new core sampler in hot cell.

FUTURE EXPERIMENTS USING THE ADAGIO FACILTY

- **High burn-up fuel**
- **Fuel subject to transient power**
- **MOX fuel**
- **Inert atmosphere**

The good results achieved this year have induced a program of analysis in order to bring new validation data for models.

The main different tests for the future are :

- tests on high burn-up fuel

- tests on fuel subject to transient power

- tests on MOX fuel: due to the MOX structure, the fuel oxidation should be different and must be studied previously

- tests in inert atmosphere: these tests are more simple than the ADAGIO experiment and some of them are already completed. They give information on gas release due to diffusion for advanced fuels.