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A New Hot Cell to Condition Highly Active Waste of Chemical Hot Cells and Robot to Condition Samples of Reprocessing Input for Analysis

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ILW coming from R&D work is solidified in a newly constructed hot cell, which is connected to the Chemical Hot Cell Facility. An commercial analytical robot has been adapted to condition dissolved spent fuel for isotope dilution analysis by mass-spectrometry

Hot Cell for ILW Solidification

Because of the varying composition of intermediate level radioactive waste originating from R&D work which is difficult to discharge to outer facilities, it was decided to construct a separate hot cell. A stainless steel cask (L = 3 m, W = 2 m, H = 2.4 m) shielded with 15 cm Pb was attached to the chemical hot cell facility and connected to the conveyor system.

The hot cell *[Fig.1]* is eqipped with three Master-Slave Manipulators, Type CRL Mg all protected by bootings and a crane of 250 kg capacity. Up to 5.10^{11}Bq (1MeV) can be handled in the cave without exceeding an outer dose rate of 5 µSv/h. The cell can be loaded by a La Calhéne DPTE 270 double lid system. The solidified waste is discharged by another double lid system of DRAHT & SCHRADER, by which a zool·drum can be loaded.

The liquid waste is neutralised prior to cementation. In 7 flasks with 7 liter capacity each, 3 to 4 l of waste solution is mixed with 5.7 kg cement.

In addition the hot cell possesses a pneumatic press to reduce the volume of emptied plastic bottles etc.

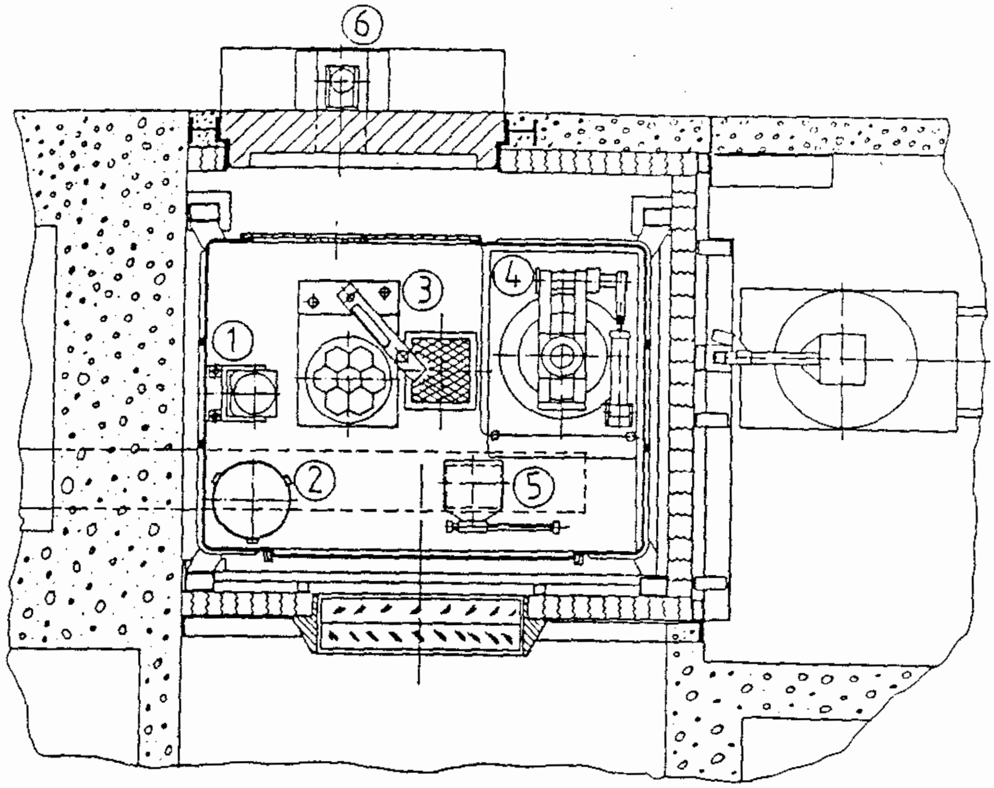

Fig. 1: *Layout of Hot Cell to solidify waste*
(1) Pneumatic Press (2) Vessel to neutralize waste solutions (3) Flasks for cementation (4) Double lid system for loading 200 l drum (5) Conveyor (6) la Calhène double lid system.

Analytical robot to condition dissolved spent fuel for analysis

The concentration of uranium and plutonium in dissolved spent fuel is generally analysed using isotope dilution mass-spectrometry. All analytical procedures developed so far consist of several steps where portions of the highly radioactive solution have to be treated: to weighed aliquots of the sample weighed amounts of uranium and plutonium spikes are added (usually ^{233}U and ^{244}Pu). To ensure a complete mixing of the Pu isotopes the valency of this element has to be adjusted carefully before uranium and plutonium are separated from fission products. The isotopic abundances of the two elements are determined by thermal ionisation mass-spectrometry complemented by alpha-spectrometry for ^{238}Pu. The tendency to burn LWR fuels up to 40 GWd/t and FBR fuels to levels twice as high and at the same time to reduce the cooling time before reprocessing from three years to one year increases the radioactivity of the sample to be analysed. Hence the occupational dose received by an analyst during conditioning of such samples for isotope dilution analysis may exceed an acceptable level if a large number of samples have to be processed.

Two measures can be taken to cope with this problem:

- reduce the sample size drastically
- condition the samples using robots to carry out the necessary manipulations

In the first case a sensitive mass-spectrometric analysis method had to be developed /1/ to allow the size of the sample to be scaled down by a factor of at least 100. The automation of the mass-spectrometric isotope-dilution analysis has been worked out before /2/ but the recent use of robots links the different steps of sample preparation more efficiently than independent automats and completely eliminates the need for the highly radioactive samples to be handled by the analyst.

In addition a method using robots completely for sample conditioning for isotope dilution analysis would allow sample treatment on-site, since the plant operator would not be involved in the preparation of the mass-spectrometer sample.

Scope of sample preparation using laboratory robots

If a robot is to carry out all steps needed to condition a sample beginning with the introduction of the sample and ending with a loaded filament for mass-spectrometry the following observations can be made:

- the robot has to be adapted to the working conditions of a glove-box
- human interference has to be minimised
- the complete process is preferably to be controlled by an expert system.

Constraints of working in a glove-box demand high reliability of the robot components and its peripheric devices. Therefore certain vulnerable parts e.g. the electronics should be separated from the instrument and installed outside the glove-box, and mechanical parts have to be protected against corrosion.

For such an automat to work unattended e.g. over night the required safety regulations have to be observed. All disposable equipment (such as test-tubes, pipettes) and the reagents have still to be loaded manually by hand into the glove-box at the same time as the sample is introduced. The disposal of the waste and remains of the sample as well as the prepared filaments for mass-spectrometry will also be carried out manually at the end of the operation.

Three pieces of information define the execution of the isotope dilution analysis:

- the identity of the sample and the required number of parallel assays
- the approximate composition of the sample
- the concentration of spike solutions available.

From this information an expert system could work out the sample size and amount of spikes to be taken by the robot, select the chemical procedure (which may differ for different fuel types) and codify the sample identity.

Setting up of an analytical robot

At the European Commission Safeguards Analytical Laboratory in Karlsruhe a commercial robot has been modified for glove-box work and is in routine use since October 1986. The robot executes the following steps:

- weighing sample and spike aliquots
- mixing and chemical adjustment of the Pu valency
- heating and drying of samples.

Since the laboratory receives samples from various reprocessing plants which reprocess different fuels, a series of programs has been set up which enables the treatment of:

- freshly diluted reprocessing input samples
- already on-site spiked samples, where only the Pu isotope mixing and exchange have to be accomplished
- preparation of dry spikes

In addition the robot is used in the analysis of Pu output samples by isotope dilution analysis.

The present set up has to be seen in connection with other automatic devices used in the laboratory where e.g. the separation of U and Pu from fission products is accomplished by automatic ion-exchange separation */3/*.

The robot is surrounded by its peripheric devices and a variety of racks for holding the samples */Fig. 2/*. The most important of these devices are balance, mixer (for liquids), liquid dispenser and heating plate */for details see fig. 2/*. The layout shows the partitioning of the box into three compartments: one used for the introduction of potentially contaminated samples, the main operation area and an area for the evaporation of liquid samples. The robot possesses two exchangeable hands, one to open and handle the possibly contaminated sample bottles and the second for the subsequent work. To avoid corrosion the robot is flushed internally with argon. Since, as mentioned above, the conditioning of the samples is plant- and fuel-dependent several standard programmes are used, which allow e.g. for different methods of isotope mixing and exchange in the treatment of the sample with NH_4F, H_2O_2, NH_2OH_2Cl.

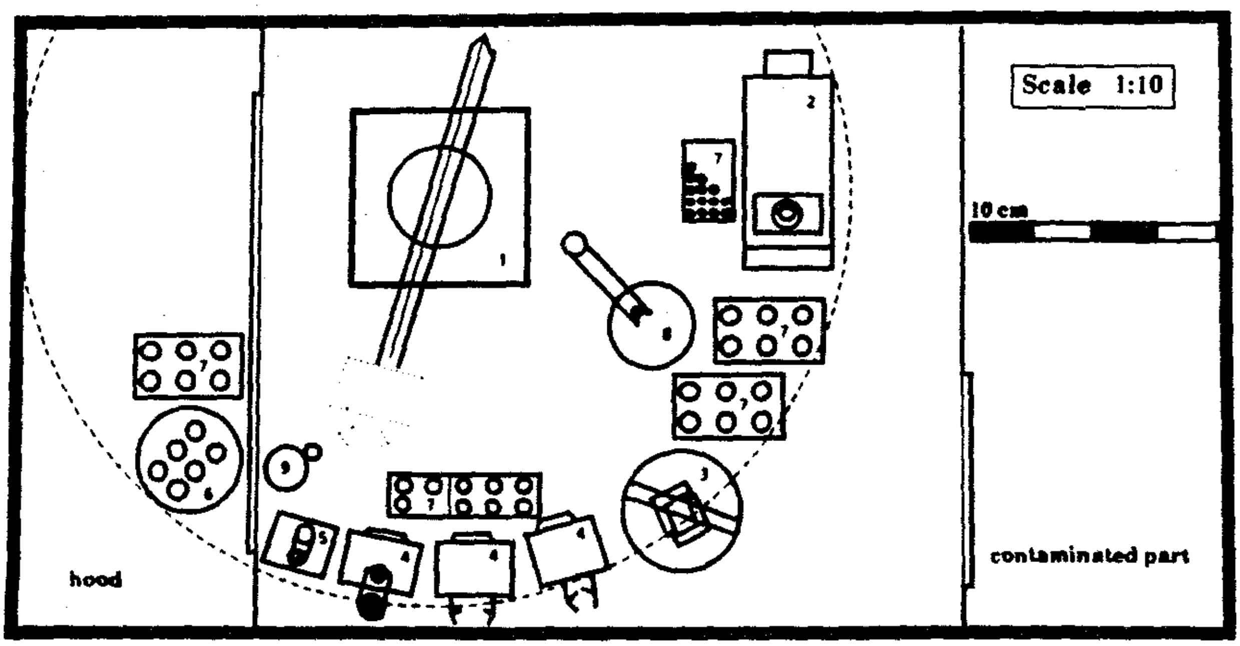

Fig. 2: *Layout of an analytical robot (1) in a glove-box, balance (2), decapper (3), exchangeable hands (4), mixer (5), heating plate (6), racks (7), waste (pipettes, etc.) (8), liquid dispenser (9).*

Performance criteria

In setting up a sample preparation system with robots the following points have to be observed:

- to ensure a steady throughput of samples a certain redundancy in equipment is needed
- quality assurance has to be implemented
- the work throughput of the robot has to be appropriate to the number of samples and/or to the capacity of the mass-spectrometer.

The reliability of the robot up to now has been extremely high and no breakdowns have been observed which could lead to a delay in the analysis. Nevertheless we feel that a standby robot is required especially when further steps of the sample preparation are executed by the robot.

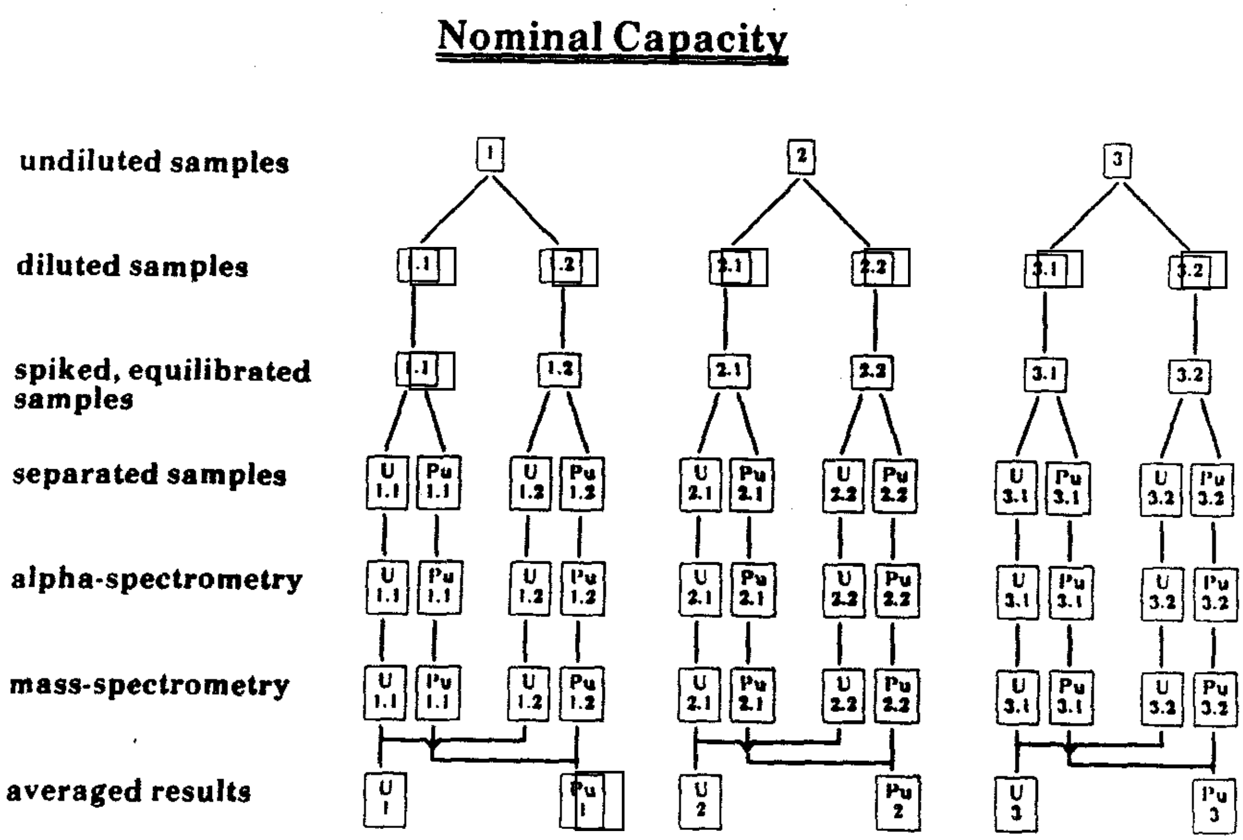

Fig. 3: Nominal capacity of the automatic laboratory

The quality assurance programme applied is not different from that used earlier in manual operation. The reproducibility of the sample preparation is controlled by duplicate analysis. The calibration of spikes and balances is routine: non-constant biases are controlled by standard weights and by analysing working standards. The latter consist of secondary standards of mixed $^{233}U/^{244}Pu$ used to monitor cross-contamination. In addition secondary standards of $(U,Pu)O_2$ are used to control the isotope dilution analysis.

The nominal capacity of the analytical robot is designed to process three input samples at the same time. From each of them two parallel dilutions are spiked and conditioned of which after separation, 12 U and Pu samples in total will be obtained *[Fig. 3]*. The mass-spectrometer turret holds 13 filaments: 12 samples and 1 isotope standard.

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