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- PROGRAMME HEVA -
MISE EN OEUVRE DU PROGRAMME INSTRUMENTATION

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- HEVA PROGRAMME -
OPERATING THE PROGRAMME INSTRUMENTATION

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1. INTRODUCTION (Plate 1)

The HEVA programme is designed to characterise the source term of fission products (FP) released by an PWR type fuel during a sequence representative of a serious accident. The tests were carried out between 1983 and 1989 and, since then, the programme has been extended by the current VERCORS programme.

This programme was operated by the Direction des Reacteurs Nucleaires du CEA and was defined and funded by the Institut de Protection et Sureté nucléaire and co-funded by EDF (French Electricity Board).

2. AIMS

The aims of the programme are as follows:

- to improve and validate models used in the ESCADRE system codes describing the risks of FPs release to the environment during a serious accident sequence,
- to create a realistic data bank for computing the behaviour of FPs in the primary circuit and the containment and, as a result, for assessing the potential release. Essentially, this could lead to the evaluation of safety margins in the light of the envelope data currently available.

Consequently, the measurements taken during the tests were aimed at characterising:

- the release kinetics and the total release of FPs and structural material as a function of fuel temperature,
- the aerosol source as a function of temperature,
- the chemical behaviour of the FPs in the fluid and in interaction with the walls.

3. EXPERIMENTAL DEVICE

The experimental device (Plate 2) includes, in the fluid flow direction :

- a supply of water vapour and hydrogen constituting the fluid in which the experiment is conducted,
- a supply of helium for protecting the graphite susceptor of the HF furnace from water vapour attack,
- a superheater to heat the fluid to 800°C,
- the test fuel, which comprises a fuel rod section from an EDF (French Electricity Board) power plant and includes three fuel pellets in their cladding. Two half-pellets of depleted uranium oxide are placed either side of the test sample and held in place by crimping the cladding (Plate 3). The sample is placed on a vertical support of dense zirconia. For the tests with the control rod material components (80% Ag, 15% In, 5% Cd alloy), the control rods were placed in a crucible positioned below the fuel rod support (Plate 4).
- the induction furnace (Plate 5), including a test channel of dense zirconia (forming a chamber that is partially sealed from the graphite susceptor protected by the slightly pressurised helium), the graphite susceptor, a double layer heat insulator (dense zirconia and alumina), a quartz tube, forming the furnace chamber, and the furnace coils,
- a junction zone with a zirconia tube internal channel,
- the cascade impactor placed in a resistance furnace with an adjustable temperature range of 250°C to 800°C,
- a heated line,
- a condenser for collecting the water vapour,
- a drier (silica gel and molecular sieve),
- a cold trap for trapping noble gases.

The entire experimental assembly is placed in a shielded cell equipped with remote manipulators, grippers and window at the LAMA, Grenoble Nuclear Research Centre hot laboratory.

The furnace is used to obtain a flat temperature profile (< 50°C) of the sample during the temperature plateau (Plate 6) and, as a result, comparable FP emissions for the three fuel pellets of the sample.

4. PARAMETER VARIATIONS AND TEST CHART

The parameters that can be varied in the HEVA tests are as follows (Plate 7):

- impactor temperature (from 250°C to 800°C),
- fluid composition and flow rate (water vapour and hydrogen),
- presence or absence of control rod components.

Only one of these parameters is varied from one test to another. The test fuel temperature is controlled by the power of the high frequency furnace. Nevertheless, the variations noted in the test chart (between 1600°C and 2100°C) are due principally to problems in adjusting the set values. On the other hand, in the VERCORS programme, the temperature is an experimental parameter.

The test chart (Plate 8) shows how the programme was implemented. It is important to note that the tests were conducted using an irradiated fuel in a PWR and that, from HEVA test 04 onwards, the fuel was re-irradiated at low power, for the sole purpose of obtaining a signal from the short lived γ emitters. HEVA test 07 was conducted with previously activated AIC components and a UO2-depleted test fuel. The programme stages were as follows:

- HEVA 01 and 02: exploratory tests with a single loop (no impactor),
- HEVA 03 to 05: study of fission product release in oxidising environment within a temperature range of 1800°C to 2000°C. Study of aerosol behaviour as a function of temperature in the 250°C to 800°C range. HEVA test 05 is the reference test for this programme.
- HEVA 06: same as HEVA 04 for the fuel temperature and HEVA 05 for the impactor temperature, but in hydrogen atmosphere.
- HEVA 07: study of behaviour of control rod components (silver, indium, cadmium) in the absence of fission products.
- HEVA 08: same as HEVA 05 with presence of AIC) (i.e., HEVA 05 + HEVA 07).

5. INSTRUMENTATION

The HEVA loop was instrumented in order to obtain the following measurements (Plates 9 and 10):

- temperatures: temperature measurement by type K thermocouples in the zones with temperatures less than 1200°C and by high temperatures thermocouples for the fuel. The important measurements are those of the superheater (TIRC3), the fuel (TIRC1 and

TIRC2), the top of the furnace (TIRC4), the junction zone (TIR4 and TIR5) and the impactor (TIR6 and TIR7).

- fluid flow rates (inlet and after the condenser).
- pressure and, in particular, the difference in pressure between the fuel rod circuit and the susceptor circuit and the pressures upstream of the impactor and the cold trap (risk of clogging).
- activity levels for which there are two types of measurement:

  . dynamic: during the test, measurements on the fuel (RIR1), the last stage of the impactor, the bead beds and the filter (RIR4).

  . static: before and after the test, measurements on the fuel and the loop components (fuel, junction zone, impactor, condensates, filters).

- aerosol size measured by the cascade impactor, dismantled after the test and analysed stage by stage by weighing and gamma-ray spectometry. The impactor was slightly modified during the course of the programme. Initially it had 8 stages (HEVA 03), this number was reduced to 6 (HEVA 04) and two bead beds were added operating in a diffusion regime (HEVA 05 to HEVA 08) (Plates 11 and 12).

  With this arrangement and in the thermohydraulic conditions of HEVA tests 05 to 08, small-sized particles could be differentiated according to their mean aerodynamic diameter (MAD) in the following way (Plate 13):

  - MAD > 0.8 μm collected in impactor,
  - 0.1 μm < MAD < 0.8 μm collected in filter,
  - 0.02 μm < MAD < 0.1 μm collected in 2nd bead bed,
  - MAD <0.02 μm collected in 1st bead bed.

- measurement of $[H^+]$ and $[I^-]$ by special electrodes placed in the condenser.

6. CLASSICAL MEASUREMENTS

These are defined as temperature, pressure and flow rate measurements.

For the temperatures, the on-line monitoring of fuel temperature and, where applicable, the temperature of the crucible containing the silver, indium and cadmium pellets is given on Plate 14, for HEVA test 08. The temperature plateau of this test was interrupted after 10 minutes because of clogging in the impactor and FP release was low.

Flow rates downstream of the condenser were measured in order to assess zircaloy oxidation kinetics by looking at the difference between the injected hydrogen and helium flow rate and the flow rate of these gases measured after condensation of the water vapour (Plate 15). In reality, small amounts of water vapour penetrated the susceptor circuit and attacked this graphite susceptor. The large volume of hydrogen generated by this reaction interfered with the measurement and the initial objective could no longer be attained. This problem is currently being solved in the VERCORS programme.

7. ON-LINE ACTIVITY MEASUREMENTS

On line activity measurements and after-test measurements using gamma spectometry are the basic measurements of the HEVA tests.

For HEVA test 08, plate 16 shows the overall variation in activity of the last components of the impactor, as a function of time, with that of the fuel temperature. It can be seen that changes in impactor activity took place almost exclusively during the temperature plateau.

Plate 17 (bottom graph) shows the variation in the fraction of cadmium released during the same test. It appears that the cadmium was released well before the temperature plateau was reached. This illustrates a problem that occurred during these tests, namely the depletion of the cadmium source before the release of the other elements and FPs. This situation is not representative of the behaviour of this element in the case of a reactor.

Plate 17 (top graph) shows the variations in the fractions of indium, iodine and caesium released. The graph shows that the release kinetics of these three elements are roughly the same.

Plate 18 (top graph) shows the same measurements made on the impactor and the bottom graph compares the kinetics of emission and arrival in the impactor of indium. It can be deduced that:

- trapping occurred in the junction zone between the furnace and the impactor (about half of the total amount emitted reached the impactor);
- element emission and arrival in the impactor were roughly synchronised (except for cadmium).

From these measurements, the release kinetics of the FPs and structural elements were calculated and compared with the envelope values currently used (Plate 19). Assumptions could then be made regarding the behaviour of certain elements. These assumptions were then tested by post-test analyses on the elements of the loop and by possible interpretations in the light of the thermohydraulic and thermodynamic data available.

8. POST-EXPERIMENT STUDIES

8.1. Test fuel

Before the test, gamma spectometry measurements were conducted on the fuel in order to draw up an initial inventory of the FPs. The value obtained was analysed using a set of modules that constitute the LINA Code used in the Laboratory. The inventory thus obtained was compared with the results obtained by the MARISE code which is used to calculate the inventory of a fuel based on its irradiation history.

The accuracy of the measurement is between 6 and 8%, and the agreement between the measurement and the calculation is always excellent (measurement/calculation ratio between 0.97 and 1.03).

For the emitting FPs, the gamma spectometry measurement is the reference value for the other measurements taken later.

After the test, the fuel was coated in situ with an epoxy resin and X-rayed (Plate 20). A gamma scan was conducted to check the overall release of each pellet (Plate 21) and fine gamma spectometry was used to measure the final FP inventory so that the percentage of FPs emitted by the fuel during the test could then be calculated.

For HEVA test 06 (in hydrogen atmosphere), gamma ray emission tomography was used to determine tellurium and barium retention in the partially oxidised cladding (Plate 22).

Finally, a macrograph of each fuel rod pellet showed the changes that had taken place during the test (Plate 23).

8.2. Composition of the loop

After the test, gamma spectometry was used to measure the loop components and a histogram was prepared to show the distribution of fission products in the components, particularly in the junction zone situated between the furnace and impactor and in the impactor itself (Plate 24). The histogram shows that a large amount of the FPs emitted was trapped upstream of the impactor, in the junction zone (about 50%).

A more detailed analysis of this junction zone showed, in particular, that the iodine and caesium were not deposited in the same way in this zone (Plate 25). This coincides with the fact that, if all the balance of the caesium was in the loop up to the outlet filter of the impactor, then the iodine progressed up to the level of the condenser in the form of a water soluble gas.

8.3. Impactor

Gamma spectometry was used to analyse the impactor components and a histogram was prepared showing the distribution of the various FPs according to where they were deposited (Plate 26).

Using the prior calibration of the impactor and taking into account the adjustments made in relation to the real conditions of the experiment, the LPMA of the Fontenay-aux-Roses Nuclear Research Centre processed the data to define the aerosol spectrum of the fission products emitted by the fuel (Plate 27).

The same procedure was followed for each FP measured and for the activation products of the control rod elements (Plates 28 and 29).

8.4. Physico-chemical analyses

The main objective of the physico-chemical analyses was to obtain data on the behaviour of the FPs in the environment in which they were released.

Thus, in the first instance, the examination covered those elements that could not be identified through gamma spectometry and with the chemical bonds linking the FPs to each other and to the other elements.

The basic method used in the HEVA programme was scanning electron microscopy combined with an analysis of backscattered X-ray energy (SEM/EDX).

This method, which is very suitable for analysing deposits, was used at the SECC, using equipment installed in a hot cell at the LAMA, and at the AEA-Winfrith laboratories, through a joint collaboration agreement for the HEVA 03 and 04 tests, and more recently for the HEVA 07 and 08 tests.

Other methods were used by the two laboratories to obtain additional information since, where chemical bonds are concerned, there is no all-embracing method that can be used. The following methods were among those used:

- X-ray photoelectron analysis (XPS), which was used, for example, to determine the chemical form of the molybdenum, the majority of which, contrary to what was expected from the thermodynamic calculations, was not bonded to the caesium,
- ionic chromatography (for element analyses in solutions after dissolving the deposits),
- infrared analysis,
- X-ray fluorescence (general and non-destructive element analysis of deposits on a surface),
- X-ray diffraction for the purpose of studying the phases present.

Whatever the method used, samples must be prepared before anlayses are conducted. For example, Plate 30 illustrates a preparation method applicable to powders deposited on the impactor plates: these are collected on a pre-glued stub with an application force monitored by means of a balance on which the impactor plate is placed. From HEVA test 06 onwards, all the solid samples were obtained in this way.

When ionic chromatography is used, a solution must be prepared beforehand. A diagram illustrating the arrangement is shown on Plate 31.

These different preparation methods are installed in a special LAMA cell.

Some results of the SEM analyses, carried out by AEA-Winfrith (HEVA 03 and 04) or the SECC (HEVA tests 05 to 08), are given in Plates 32 to 35.

9. SOME RESULTS

By way of an example, some results corresponding to the predetermined objectives of this programme are given below.

9.1 Release rate coefficients

The release rate coefficients are defined by the equation:

$$F\ (min^{-1}) = \frac{Log\ (1/1\ -FR)}{t}$$

where :

t = duration of plateau (in mins.) at the temperature of the test,
FR = inventory fraction released during the test.

The HEVA programme tests show that (Plate 36):

1) in an atmosphere of water vapour and hydrogen (oxidising environment), the FP release rate coefficient values were as follows in relation to the envelope values given in CORSOR 1:

   Cs, I, Xe, KR : lower by a factor of 3

   Sb : higher by a factor of 3

   Mo: higher by a factor of 5 to 10

   Ba: roughly the same

   Ru, Zr: higher by a factor of about 10.

2) in a hydrogen atmosphere, compared with the previous values:

Cs, I: lower by a factor of 3

Te, Mo: lower by a factor of about 10

Ba: higher by a factor of about 10.

3) there is no indication of the influence of the control rod components. This conclusion is not final insofar as some of these products were emitted before the FPs were emitted.

9.2. Aerosol size (Plate 37)

At 800°C the FPs are in vapour form and at 250°C the mean aerodynamic diameter of the aerosols is less than 0.2 mm. The possible influence of the nature of the atmosphere or the control rod components was not shown.

9.3. Chemical forms

Information concerning the chemical forms detected is as follows:

Cs: mainly CsOH which transforms into silicates or carbonates,
I: not deposited in the same places as the Cs,
Mo: mostly MoO_3,
Sn: SnO_2,
U: $UO_{2.25}$ (over-stoichiometric composition).

For the tellurium, it is assumed that there was an interaction with CsOH and the tin, even though it is not shown.

10. CONTINUATION OF PROGRAMME

The HEVA programme has been extended by the VERCORS programme in which two tests have been conducted. The main objective of this programme is to study the release of non-volatile FPs up to the fuel fusion stage. Based on what was learned during the HEVA programme, the tests will be conducted at a higher temperature (2400°C) in an initial stage and then, if possible, up to fuel fusion. To achieve this objective, a new loop must be created, in a new environment and with new instrumentation. The list of improvements is shown on Plate 38.

1) loop modifications:

- study of virtually non-volatile fission product release: development of a very high

temperature furnace,

- development of a new fluid injection system,
- installation of a new circuit downstream of the furnace.

2) Improvements to instrumentation

- high temperature measurements: radiation pyrometry,
- study of zircaloy oxidation: gas-chromatography,
- study of noble gas release: gamma spectometry
- study of vapour phase deposition: development of a temperature gradient tube. This study is backed up by the analytical tests conducted in the DEVAP programme.
- improvement of iodine chemistry: specific filter.

Work has started on these improvements and they should be operational by the beginning of 1994 when the new VERCORS cell is commissioned (Plates 39 and 40).

3) Improvements in post-experiment analysis systems

- development of selective dissolution methods,
- improvements in ion chromatography,
- greater use of methods to reveal chemical forms.

4) Improvements to injection system of AIC control rod components

- injection of components in the fluid in the form of controlled flow vapour. This study is supported by the EMAIC programme tests.

5) Programme support

- DEVAP: programme to study vapour phase deposits of volatile FPs. It takes place in a special loop and the objectives are as follows:
  - to provide FP deposition rates on surfaces representative of those of the main constituents of a primary circuit,
  - to provide information on FP distribution between vapour deposits and particles,
  - to study the feasibility of and then interpret the deposition section planned for the VERCORS tests.
- EMAIC: programme to study the emission of components of an AIC rod after cladding failure. The main aim of the programme is to provide emission kinetics values for the components in order to:

- inject them in the VERCORS programme with correct representativity,
- integrate them in the release computation modules when the core is destroyed.

11. CONCLUSIONS

This programme represents another step foward in the knowledge of the source of FPs released by the fuel during a serious accident sequence and will allow increasing the accuracy of the source term computing with the results of the measurements of :

- the kinetic of the volatile elements release (Cs, I, Xe, Kr) in an oxidising atmosphere : lower by a factor of 2 to 3 in relation to the envelope values,

- the kinetic of the same elements in a reducing atmosphere : lower by a fact-or of 3 in relation to the previous values,

- the aerosol source size near the fuel with a mean mass aerodynamic diameter lower than 1 μm.

The full extend of this advance will be felt when, in cooperation with scientific laboratories throughout the world, this programme is carefully compared with others with similar objectives that are either being developped or have been completed.

This experimental results were obtained using the abilities and the means developped in the Service d'Etude du Comportement de Combustible of the Nuclear Research Center of GRENOBLE for the tests operating and the fission product measurements.