9.1 Current Status of VEGA Program and a Preliminary Test with Cesium Iodide

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Abstract

The VEGA program has been performed at JAERI to clarify the mechanism of FP release from irradiated PWR/BWR fuels including MOX fuel and to improve predictability of the source term. The principal purposes are to investigate the release of actinides and FPs including non-volatile radionuclides from irradiated fuel at 3000 ℃ under high pressure condition up to 1.0 MPa. The short-life radionuclides will be accumulated by re-irradiation of test fuel just before the experiment using the JAERI's research reactor such as JRR-3 or NSRR. The test facility was installed into the beta/gamma concrete No.5 cell at RFEF and completed in February, 1999. Before the first VEGA-1 test in September, 1999, a preliminary test using a cold simulant, cesium iodide (CsI) was performed to confirm the fundamental capabilities of the test facility. The test results showed that the trapping efficiency of the aerosol filters is about 98 %. The amount of CsI which arrived at the downstream pipe of the filters was quite small while a small amount of I_2 gas which can pass through the filters was condensed just before the cold condenser as expected in the design.

Key Words: *Severe Accident, Radionuclide Release, VEGA Program, Non-Volatile Fission Products, Source Term, Cesium Iodide, I_2 gas, Aerosol, Condensation, Deposition*

1. INTRODUCTION

The release of radionuclides such as fission products (FPs) and actinides from fuels is one of key issues for precisely estimating the source terms in hypothetical severe accidents[1]. A lot of experimental researches such as ORNL/HI-VI (USA)[2], CEN Grenoble/HEVA-VERCORS (France)[3] and CRL/HCE-HCL (Canada)[4] tests have been conducted so far. However, the number of the tests and the covering test conditions were limited. Thus, considerable uncertainties still remain, especially in the areas under high temperature and/or high pressure conditions and the behavior of short half-life FPs. For example, the ambient atmosphere such as oxidizing or inert condition could affect the chemical form of released radionuclides which depends on the volatility. The bundle material such as control rod could react chemically with the released radionuclides and could change their volatility. The FP release from very high burnup fuel could be enhanced compared with that from low burnup fuel. The FP release rate could become small as the pressure increases[5]. There is no data from MOX (mixed-oxide fuel).

Moreover, recent VERCORS tests[6] showed that Sr is less volatile than Ba although the both elements were classified in the same group in the CORSOR-Booth model[7]. Moreover, it was pointed out that large amount of Ba release would result in approximately 20% decrease in residual decay power of degraded core during severe accidents[8]. Accordingly, the Ba release behavior could delay the accident progression as well as the change of FP source term.

In order to solve these uncertainties and to investigate the releases of FPs and actinides from irradiated fuel at 3000 ℃ under high pressure condition up to 1.0 MPa, an experimental program, VEGA (Verification Experiments of radionuclides Gas/Aerosol release) has been performed at Japan Atomic Energy Research Institute (JAERI)[9),10),11]. The summary of previous studies and target of VEGA program are shown in **Table 1**. In the VEGA program, a first priority is made for the release of volatile/non-volatile FPs and actinides under high temperature above 2700 ℃ and high pressure conditions. One of special features of this program is to investigate the effect of ambient pressure on the radionuclide

release from fuel which has never been examined in previous studies. In the experiment, the irradiated PWR/BWR fuels including MOX will be used as the test sample. The other objective of this program is to investigate the effect of oxidized/unoxidized conditions on the radionuclide release/transport behavior in reactor core and coolant systems.

2. OUTLINES OF VEGA FACILITY

The test facility was installed into the beta/gamma concrete No.5 cell at the Reactor Fuel Examination Facility (RFEF) and completed in February, 1999. Schematic of VEGA facility is shown in **Fig.1**. The facility mainly consists of a steam/gas supply system, high-frequency induction furnace, thermal gradient tubes (TGTs), aerosol/charcoal filters, cascade impactor and on-line gamma measurement system. The test section is pressurized up to 1.0 MPa between the gas supply system and the valves just after the aerosol filter. During the experiment, one of three valves will be opened depending on the fuel temperature. The valve of cascade impactor line will be opened for several minutes to measure the aerosol size distribution. The temperature of piping between the furnace and TGTs is maintained at 750 ℃ to avoid CsI aerosol deposition. The temperature at TGTs is decreased linearly from 750 to 200 ℃ to obtain the FP deposition data as a function of temperature and to identify the chemical form of deposited FPs.

Depending on the maximum temperature of induction furnace and the oxidized/unoxidized carrier gas conditions, the materials used in the VEGA furnace will be changed as shown in **Table 2**. In the case of the maximum temperature below 2200℃, ZrO_2 will be used as crucible materials. In the case of the maximum temperature above 2200℃ and unoxidized condition, tungsten (W) will be used while ThO_2 will be used under oxidized condition.

The latest design of furnace for oxidized condition is shown in **Fig. 2**. The induction coil is located just outside of alumina tube. Only the material which has an electric conductivity in circumferential direction can be heated by induction coil. In this design, graphite cylindrical susceptor can be heated. The inner tube is a pressure boundary and there is no steam flow near graphite susceptor to avoid oxidation of graphite. The furnace is surrounded by thermal insulator made of carbon felt and carbon powder. Temperatures of furnace are measured by two pyrometers. One pyrometer measures temperature at crucible bottom while the other one measures the side of susceptor as the backup. In order to keep gas flow path, lateral holes were made on the stand pipe and crucible. Moreover, a cap was covered above crucible to help carrier gas to come into crucible and to sweep released FPs. The fundamental design of W furnace for unoxidized condition is basically the same as that of ZrO_2 or ThO_2 furnace. One of special features of W furnace is that there is no graphite susceptor and instead W inner tube is directly heated by induction coil.

The fabrication study of ThO_2 tubes is being performed. Since ThO_2 is a nuclear material, the handling is not so easy. JAERI has conducted the fabrication of ThO_2 tube by using the centrifugal slip casting, rubber pressing or metal mold techniques. Recent study showed that the ThO_2 powder size growth (about 1 μ m) by heating up to 1,500 ℃ is very important for improvement of the mold strength.

Photograph 1 shows the VEGA facility taken through a lead grass of concrete cell. The furnace and induction coil are located inside the right-hand chamber. **Photograph 2** shows the gaseous FP trap device which is set at the isolation room next to the hot cell. The carrier gas including released FPs enters, at first, condenser and gaseous iodine is removed by charcoal at 0℃. Then, the carrier gas is delivered to dryer and noble gas trap and the noble gas is removed by charcoal cooled by Fluorinert (compound of fluorine) less than -70℃. Emitted gamma rays from the inside condenser and noble gas trap are measured by the Ge semiconductor detectors.

3. EXPERIMENTAL CONDITIONS

The maximum size of test sample is 6 cm long and the diameter is 14.3 mm. The weight of test sample with or without cladding is about 100 g. The samples used in the VEGA experiments are irradiated PWR, BWR or MOX fuels of which burnup varies from 26 to 61 GWd/tU. In order to accumulate the short-life radionuclides such as I-131, re-irradiation of test fuel just before the experiment will be done using JAERI's research reactor such as JRR-3 or NSRR (Nuclear Safety Research Reactor). The maximum temperature of

test fuel can be raised up to 3,000 ℃ and the ambient pressure will be changed from 0.1 to 1.0 MPa. In order to investigate the effect of ambient gas species on the FP release behavior and the chemical form of released FPs, helium, hydrogen, air, steam or the mixed gas of them will be used. Moreover, the effect of control rod materials (such as Ag, In, Cd and boron) or the fuel burnup on the FP release behavior will be examined.

4. MEASUREMENTS AND CHEMICAL ANALYSES

The transport and deposition behaviors of released FPs will be studied by chemically measuring the deposition masses onto pipings such as TGTs under well characterized flows of steam/H_2/He/air atmosphere monitored by on-line oxygen/hydrogen sensors. On-line gamma measurement will be done for aerosol filters, cascade impactor, charcoal and cooled charcoal traps. Radionuclides such as Cs-137, I-131, Ba-140 and La-140 will be trapped and measured at aerosol filters and cascade impactors. Gaseous I-131 will be captured at charcoal trap. Noble gases such as Kr-85 and Xe-133 will be trapped at cooled charcoal trap. On-line measurement of O_2/H_2 concentrations will be done at just after aerosol filters. Off-line gamma spectrometry will also be performed after the experiment for most of downstream pipings including crucible in furnace.

The analyses with SEM/EPMA (Scanning Electron Microscope/Electron Probe X-Ray Micro Analyzer) will be performed to identify elements deposited onto piping. The analyses with SIMA (Secondary Ion Micro Analyzer) and ICP-AES (Inductively Coupled Plasma Atomic Emission Spectrometer) will also be conducted for measurement of inorganic materials in solution.

The radioactivity of various nuclides calculated by the ORIGEN-2 code[12] is shown in **Table 3**. The calculated fuel is PWR MOX of which weight is 104 gUO_2 and its burnup is 56 GWd/t. The cooling duration is 1 year. The reason for use of this PWR MOX in the calculation is that radioactivity becomes the highest among expected candidates for fuel specimens. It was also assumed that NSRR re-irradiation with linear heating rate of 0.065 W/m is performed for 9 hours and after that, the sample is cooled for 3 days. Kr-85 and Cs-137 will not be accumulated in sample fuel pellet by NSRR re-irradiation because their yield mass are small. On the other hand, relatively large amount of short half-life radionuclides such as I-131, Xe-133m, Ba-140 and Np-239 will be accumulated and their radioactivities will become large enough for measurement.

5. TEST MATRIX

Preliminary VEGA test schedule is shown in **Table 4**. The shakedown of test facility was finished by the beginning of FY1999 and first VEGA-1 test was performed in September 9, 1999. The maximum temperature of experiment will be gradually increased. The re-irradiation of test sample using JAERI's research reactor will be done for one experiment a year. Three experiments a year are scheduled after FY2000. In near future, radionuclide release from MOX will be investigated. This is because there is a difference in yield-mass between U-235 and Pu-239. Moreover, plutonium is very poisonous compared with uranium.

6. PRELIMINARY TEST WITH CESIUM IODIDE

Before the VEGA-1 test, a preliminary test using a cold simulant, CsI instead of irradiated fuel was performed to confirm the fundamental capabilities of the test facility[11]. Prior to the CsI test, several coupons were set at three different locations to measure CsI deposition onto them. The location of SUS316 coupons for CsI test is shown in **Fig. 3**. It is noted that two coupons were set at the same place to confirm reproducibility of measurement. Moreover, aerosol deposition onto 4 inner SUS tubes which are inserted in TGTs were measured. Again, by measuring CsI deposition at different places inside VEGA facility, an attempt was made to obtain trapping efficiency of aerosol filter and to confirm almost no arrival of CsI aerosol at condenser as expected in the design.

The furnace temperature of CsI test is shown in **Fig. 4**. Temperature was increased at a rate of about 1 ℃/s. A small plateau was made at 1,350℃ because the pyrometer had to be changed from monochromatic to two color for higher temperature measurement. After that, temperature was raised and maintained at 1,500℃ for 30 min and was decreased mostly in the same manner. The deposited mass distribution and

temperature at 60 min are shown in **Fig. 5**. The temperature simply decreased as the distance becomes far from fuel.

The cesium and iodine masses deposited onto the coupons were measured separately by using the ion meter and the liquid ion chromatography. The deposition masses of cesium and iodine increased at downstream of TGTs and decreased by two orders of magnitude at filter. This result indicates that the trapping efficiency of filter is about 98 %. Moreover, deposition masses of cesium and iodine were almost the same at the piping between TGTs and filters. Therefore, it can be said that most of CsI was vaporized, transported and deposited as the original chemical form of CsI.

However, there was an obvious difference in deposited mass just before condenser between cesium and iodine. This is considered that small amount of CsI was dissolved due to impurities such as humidity and formed I_2 gas which can pass through the filters was condensed just before the cold condenser. However, these results indicates that any aerosol did not arrive at condenser as expected in design. The test results also showed that a relatively large amount of CsI may be deposited at the outlet piping of furnace in the case that the flow rate is not kept high enough. Since the gamma measurement at that portion is difficult for present design due to its structural reason, the design will be changed in future experiments.

7. CONCLUSIONS

The VEGA program has been performed at JAERI to clarify the mechanism of FP release from irradiated PWR/BWR fuels including MOX and to improve predictability of the source term. The principal purposes of this research are to investigate the release of actinides and FPs including non-volatile radionuclides from irradiated fuel at 3000 ℃ under high pressure condition up to 1.0 MPa. The test facility was completed in February, 1999. Before the first VEGA-1 test in September, 1999, a preliminary test using a cold simulant, CsI instead of irradiated fuel was performed to confirm the fundamental capabilities of the test facility. The test results showed that the trapping efficiency of the aerosol filters is about 98 %. The amount of CsI which arrived at the downstream pipe of the filters was quite small while a small amount of I_2 gas which can pass through the filters was condensed just before the cold condenser as expected in the design.

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Table 1 Previous studies and target of VEGA

Parameters / Radionuclides	＜2700℃	≧2700℃	Pressure	Ambient gas
Volatile FP	○	×	×	△
Non-volatile FP	△	×	×	△
Actinide (Mox fuel)	△	×	×	×
Short-life FP	△	×	×	×

○ ;Sufficient data, △ ; Insufficient data, × ; No data

First priority of VEGA　Second priority of VEGA

Table 2 Requirement for furnace material

	M.P. > 3000℃	Stability under oxidizing condition
ThO_2	○ (3370℃)	○
W	○ (3382℃)	×
ZrO_2	× (2677℃)	○

Table 3 Gamma measurement

(ORIGEN-2 calculation)

Nuclide	E-gamma (MeV)	Half life (day)	I-gamma (%)	LWR*1) (Ci)	Re-irradiation in NSRR*2) (Ci)	Total (Ci)
Kr-85	0.514	3912.8	0.434	8.91E-01	-	8.91E-01
I-131	0.364	8.04	81.2	-	8.07E-02	8.07E-02
Xe-133m	0.233	2.19	10.3	-	1.32E-02	1.32E-02
Cs-137	0.662	10950	85.2	1.74E+01	-	1.74E+01
Ba-140	0.537	12.75	24.39	4.99E-02	1.21E-01	1.71E-01
Np-239	0.278	2.36	14.1	-	1.00E-01	1.00E-01

*1) PWR MOX fuel (104gUO_2, Burnup; 56GWd/t, Cooling duration : 1 year)

*2) NSRR re-irradiation ; 0.065W/m for 9hrs and cooling for 3days

Table 4 VEGA test schedule

	1999	2000	2001	2002	2003	2004	2005
ThO_2 tube fabrication							
Shakedown							
Inert condition							
Oxidizing condition							
Pressure effect (+burnup)							
Short-life FP							
MOX fuel test							

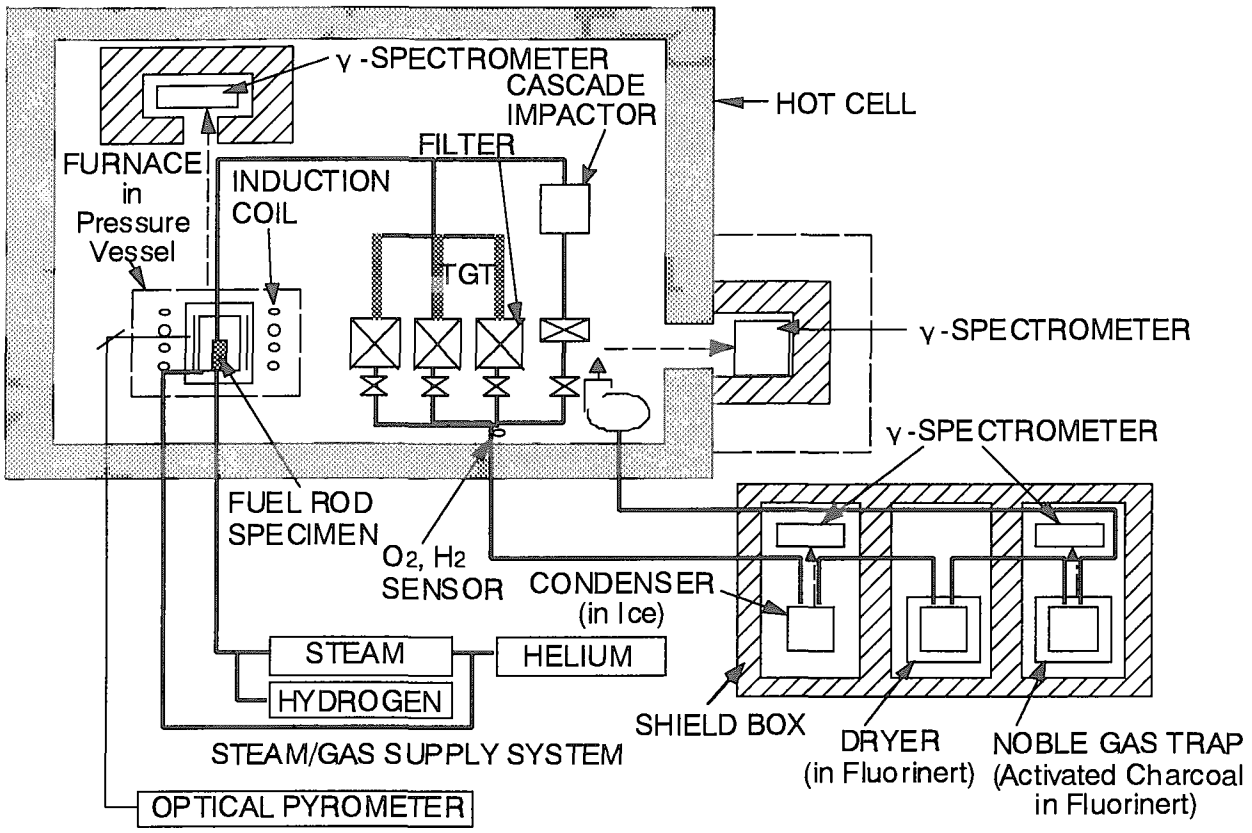

Fig.1 Schematic of VEGA facility

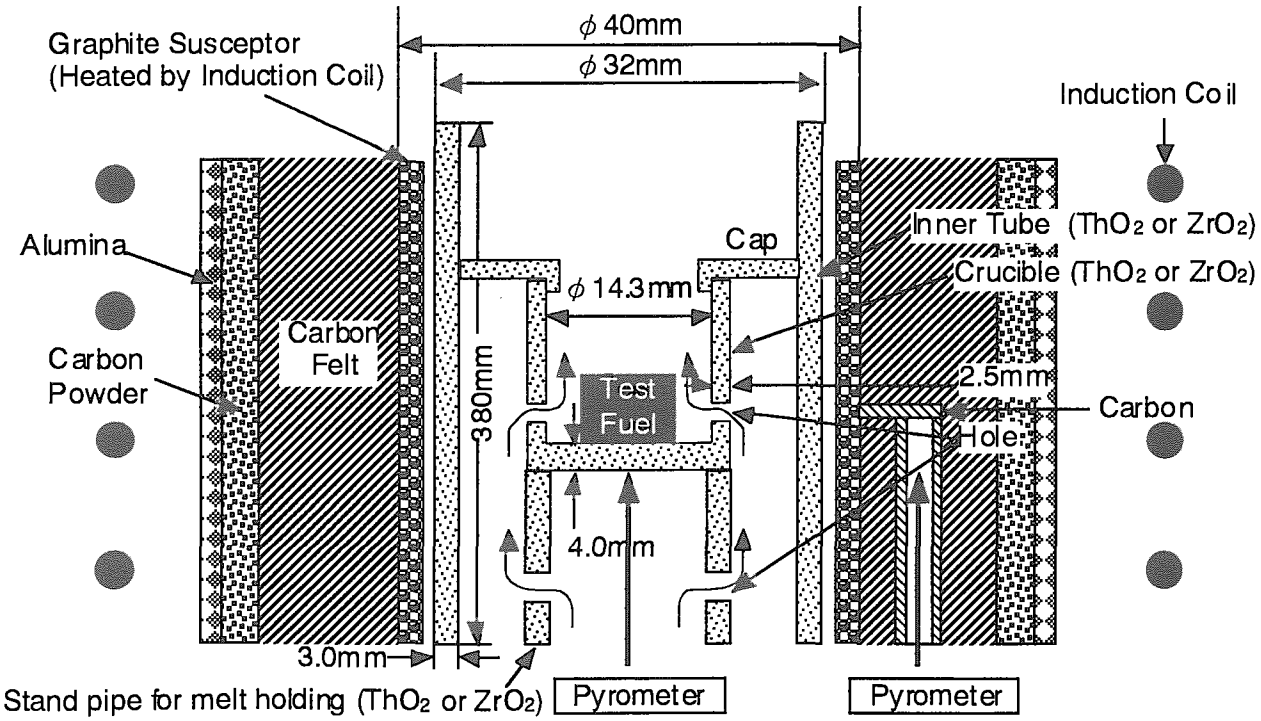

Fig.2 VEGA furnace for oxidized condition

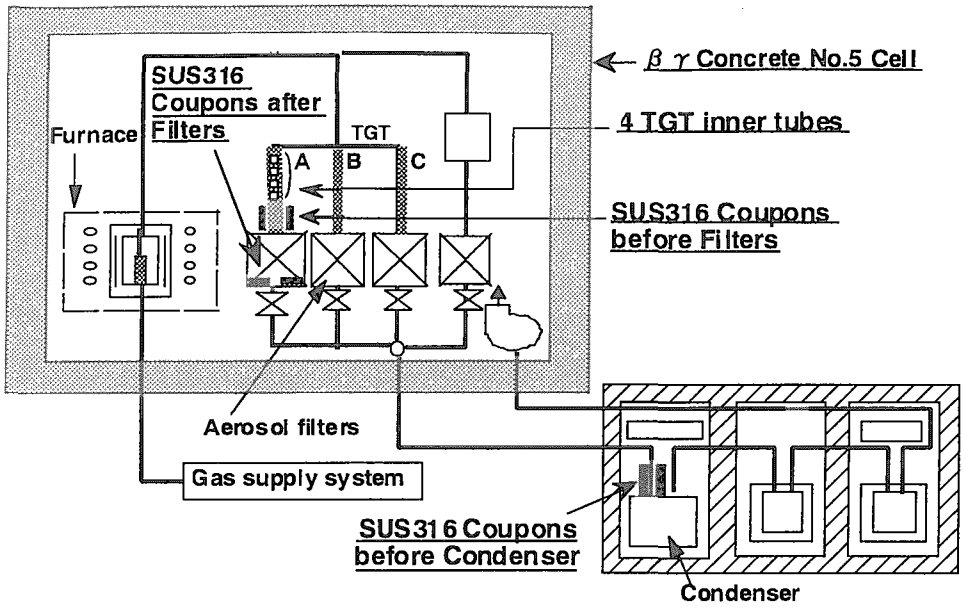

Fig.3 Location of SUS316 coupons for CsI test

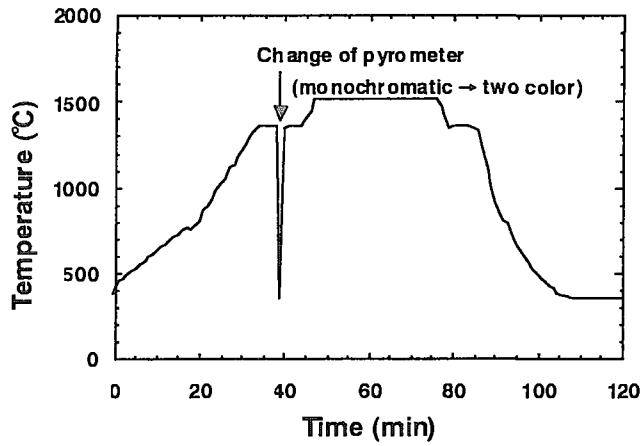

Fig.4 Furnace temperature of CsI test

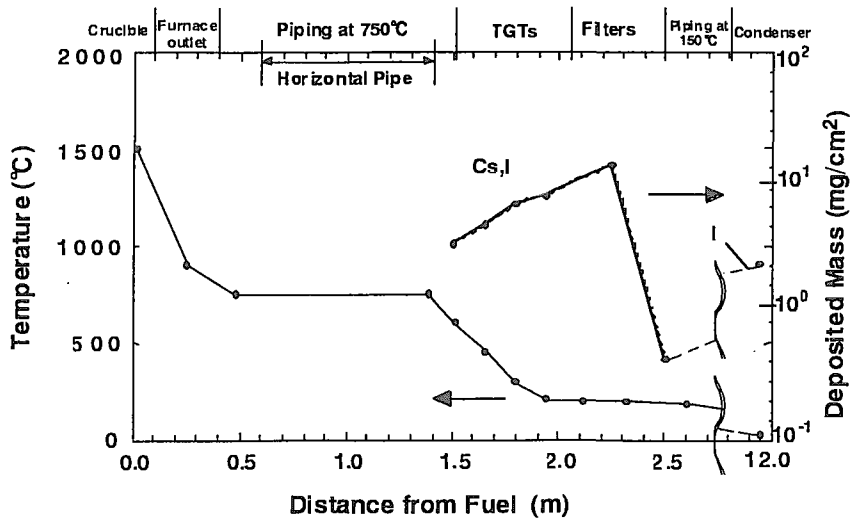

Fig.5 Deposited mass and temperature at 60 min

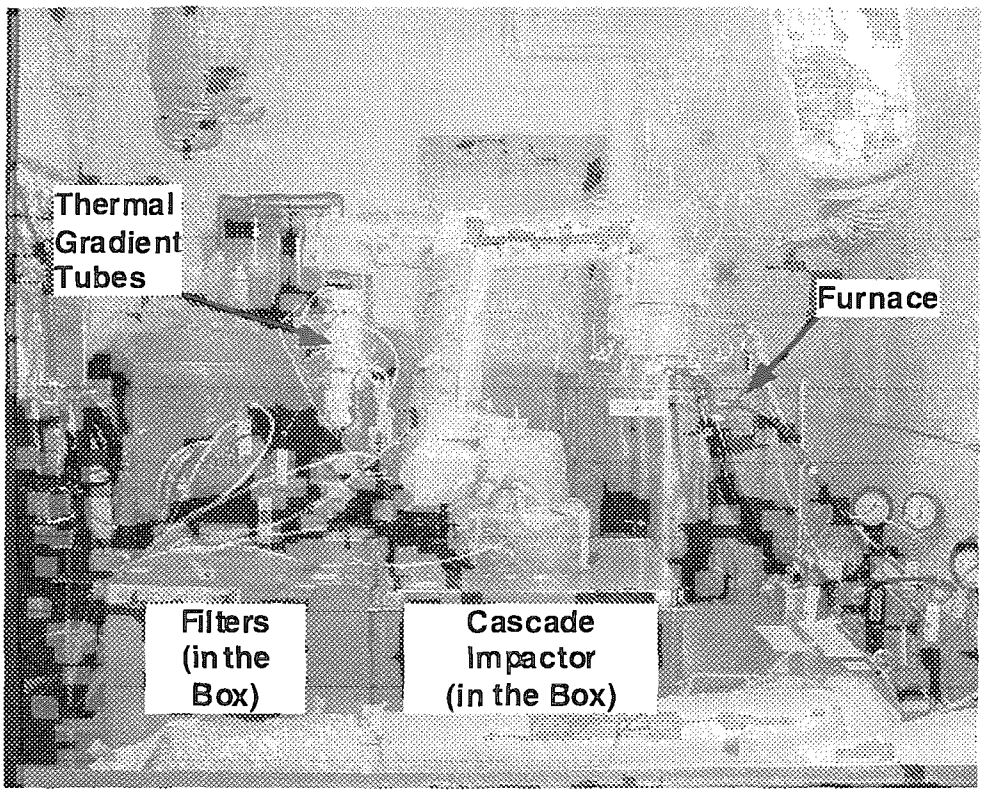

Photo.1 VEGA Facility

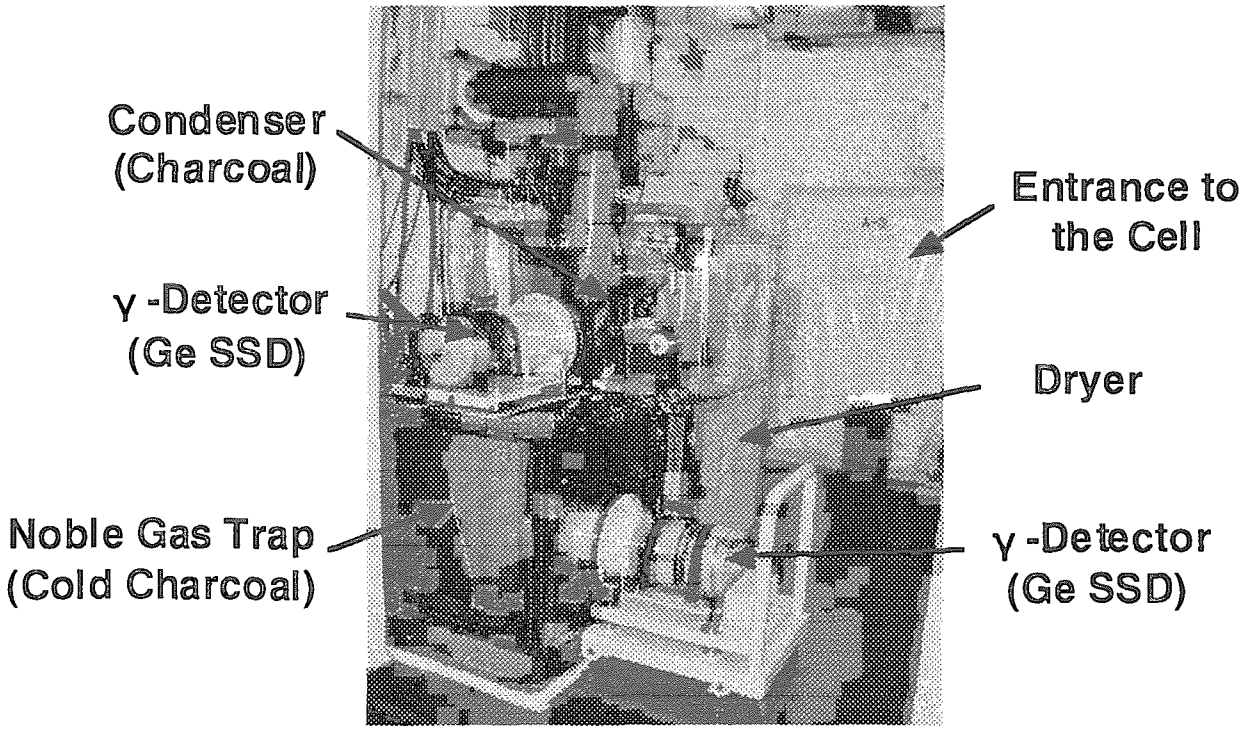

Photo.2 Gaseous FP trap device