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SAFETY ASPECTS OF LWR FUEL REPROCESSING AND MIXED OXIDE FUEL FABRICATION PLANTS

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1. INTRODUCTION

The overriding issue involved in the use of nuclear energy is to protect the public against the potential hazards from the operation of reactors and the supporting fuel cycle facilities. On the basis of rigorous evaluations of the safety, environmental and health effects for each segment in the fuel cycle it must be demonstrated that the risks remain at generally acceptable low levels for both normal and accidental conditions.

This paper discusses certain aspects of the safety of large fuel reprocessing and mixed oxide fuel fabrication plants during both normal operation and postulated accidents. The accidents reviewed in the paper are of the serious types that realistically can be postulated or have occurred.

Because of basic functional differences between reprocessing plants, fuel fabrication plants and nuclear power reactors, the structure and safety systems of these plants are different in many respects. The most important differences that influences safety systems are:

- Both fuel reprocessing and fabrication plants do not have the high system pressure that is associated with light water reactors.
- A considerable amount of the radioactivity of the fuel, which is in the form of short lived radionuclides has decayed before reprocessing.
- Most of the fission product inventory accumulated in a reprocessing plant (except for the fuel storage pool) is in the liquid state in constrast to reactor conditions.
- In spite of the degree of automation already achieved, Plutonium fuel manufacturing still implies the largest fraction of direct human access to pure fissionable material in the whole fuel cycle.

Therefore, fuel reprocessing plants and mixed oxide fuel fabrication plants are designed - like power reactors - with multiple confinement barriers for control of radioactive materials, but do not require the high-pressure components and containment systems that are used in LWR's.

The safety technology of each segment in the fuel cycle will be measured by comparing with the very high standards developed for LWR's. The outstanding success in reactor safety /1/ is based on the "defense in depth" approach.

Therefore, the same basic safety approach is applied for reprocessing and fuel fabrication plants taking into account their specific characteristics and differences.

2. BASIC APPROACH AND PRACTICES FOR ASSURING SAFETY

The potential hazard involved in reactor and supporting fuel cycle facilities requires strict criteria and comprehensive safety evaluation for licensing actions. Therefore the following major approach is applied to assure public safety. Multiple confinement barriers are used against the escape of radioactivity from nuclear facilities. The practices to assure that the function of these barriers and related safety systems will not be impaired also under accident conditions is called a "defense in depth" design. Briefly, this concept can be summarized by the following "three levels of safety" /1, 2, 3/:

1) Prevent accidents through designing for maximum safety in normal operation and maximum tolerance for system malfunctions. Use design features inherently favorable to safe operation. Emphasize quality assurance, redundancy, inspectability and testability.
2) Assume incidents will occur in spite of care in design, construction and operation. Provide saftey systems to protect operators and the public and to prevent or limit consequences of such incidents to tolerable levels.
3) Provide additional safety systems against hypothetical accidents, where some protective systems are assumed to fail simultaneously with the accident they are intended to control.

This safety concept has been proven to be very successfully on the basis of about 45o reactor years all over the world without an accident affecting the public.

With respect to fuel reprocessing and fabrication plants only, the statistical evidence from the small number of documented laboratory and plant incidents is up to now not sufficient to demonstrate the success of that safety philosophy in these facilities. Therefore detailed analysis of potential accidents have to be extended concerning both probability and consequences for cases which can be realistically postulated.

However, safe operation with smaller reprocessing plants were already demonstrated, as for example the Eurochemic plant /4/ and the very successfully working pilot reprocessing plant "Wiederaufarbeitungsanlage Karlsruhe WAK" /5/.

3. SAFETY ASPECTS OF REPROCESSING PLANTS

3.1 Process description from a safety point of view

In Fig. 1 a scheme of the principal process steps is shown. The following process steps are of main interest from the safety point of view:

- Unloading of fuel cask and storage of fuel elements
- mechanical handling of fuel elements, chopping and dissolution
- 1. solvent extraction cycle and plutonium purification cycles
- treatment and storage of fission product solutions
- treatment and storage of medium active waste.

The radioactive inventories to be considered here are based on a large reprocessing plant with a capacity of 15oo t U per year and a peak daily throughput of about 8 t U. The following data of the activities are calculated with the ORIGEN-code /6/ for a fuel with a burn-up of 4o GWd/tU and 3,2 % U 235 enrichment.

When discharged from a light water reactor the total fission product content amounts to about 1,9 x $1o^8$ Ci/tU. The decrease of this activity with time is shown in Fig. 2. After a cooling time of 200 days the activity has decreased to about 5 x $1o^7$ Ci/tU at the time of delivery to the reprocessing plant.

In view of the large buffer capacity of the storage facilities the average cooling time will be approximately 6oo days, at which time the activity has further decreased to about 2 x 10^6 Ci/tU. The activity content in the process building itself is much lower and in addition distributed over a large number of cells, Fig. 3. The bulk of activity is concentrated in the fuel storage ponds and the high level waste storage tanks. A typical fuel storage facility, containing a fuel inventory of the process capacity per year of the reprocessing plant (15oo t) would thus roughly contain an amount of activity equivalent to one pressurized water reactor of the 1300 MWe class after 2,5 years full power operation /7/. The isotopic composition of this activity is of course different from the one in the reactor.

In the first extraction cycle the main bulk of fission product activity is separated from Uranium and Plutonium. After concentration the fission product solutions are stored. Assuming a five year cooling time before solidification, fission products from 7500 t Uranium will be accumulated. They contain 6×10^{8} Ci of Sr 90, 9×10^{8} Ci of Cs 137 and in addition other radioactive isotopes up to 5×10^{9} Ci depending on the total cooling time.

Accumulation of larger amounts of Pu will be avoided by direct transfer of the Pu nitrate product to the fuel fabrication facility. The fuel fabrication plant will contain the main amount of the plutonium totally involved.

The Uranium product does not pose safety problems relative to Pu and fission products.

3.2 Normal operation

3.2.1 Formation of airborne activity

Only a negligible amount of airborne activity will be released from the discharge and storage operations due to the integrity of fuel canning, and due to the fact that defective fuel elements are stored in tight casks.

When the fuel is exposed during chopping and subsequent dissolution a major part of the volatile radioactive isotopes, namely Kr-85, I-129, C-14 are liberated and enter into the dissolver off-gas cleaning system.
But only a smaller fraction (approx. .5 %) of Tritium (H-3) is released to the off-gas cleaning system.

From the solutions liquid aerosols are formed during transfers with airlifts and steam jets or by air sparging and other processes. The nuclide composition in the aerosols is the same as that in the liquid of origin and will not change considerably in the time period of interest.
Due to existing technology for the retention of radioactive isotopes (see 3.2.2) effluents also from large fuel processing plants can be kept sufficiently low so that doses in the environment stay below permissible limits.

3.2.2 Off-gas cleaning

Regulatory requirements in the Federal Republic of Germany limit the whole body dose to 30 mrem/a and the thyroid dose to 90 mrem/a.

Effective means for the retention of the radioactivity in vessel off-gases are available in order to keep the radiation dose below the specified limits. Regarding the dissolver off-gas cleaning system technological improvements and demonstration are required. A concept of the dissolver off-gas cleaning system is shown in Fig 4. This off-gas cleaning system for both normal and accident conditions is in an advanced development stage in the Kernforschungszentrum Karlsruhe /8,9/. On the basis of cold engineering tests, a demonstration pilot off-gas cleaning system will be constructed and operated in connection with the WAK plant. Using prototypic components of a large 1500 t/a reprocessing plant, it will be demonstrated that the efficiency and availability of such a retention system for α-aerosols, I, NO_x and Kr meet the requirements /10, 11/. The removal of Kr 85 from the dissolver-off-gas can be achieved by low temperature rectification, including a precleaning process step to separate impurities.

Retention of iodine in the dissolver off-gas has been demonstrated technically at the WAK. Retention efficiencies better than 99,9 % have been achieved /12/.

Aerosol activity can be effectively retained by high efficiency air (HEPA) filters which are well proven in nuclear technology. Their performance can be further improved by using demisters to reduce liquid aerosol particles. The use of electrostatic filters has also been demonstrated successfully for vessel off-gas cleaning whereas such devices promise little advantages in the dissolver off-gas cleaning system

3.2.3 Environmental effects

With the special analysis of the environmental effects of Plutonium- recycling in LWR's the GESMO Report /13/ closed a gap within the series of environmental analysis reports concerning the radiological impact of nuclear industry.

The most important prerequisite of the analysis of the radiological impact resulting from a large reprocessing plant is the knowledge of, or realistic estimates about release rates of radionuclides. The release rates are based on the decontamination factors involved in the retention and cleaning systems. Considerable development efforts are in progress in the Federal Republic of Germany /14, 15/ to assure that advanced retention and off gas cleaning systems for Kr-85, I-129 and α-aerosols will be available for large reprocessing plants.

With regard to regulatory requirements /16/ the release rate for Kr-85 from a large reprocessing plant should not exceed 10^6 Ci/a Kr-85 /17/. Corresponding figures of I-129 will be in the range of a few hundred mCi/a.
This corresponds to retention factors of 90 % for Kr-85 and 99 to 99,8 % for I-129.

The release fraction of Pu isotopes, Am-241 and -243, Cm-242 and -244 as compared to the total plant inventory of these nuclides can be kept as low as 10^{-7} to 10^{-8} with one barrier of absolut filters which is sufficient in small plants. The corresponding requirements for large plants have been determined to 10^{-8} /18/ which canbe met without difficulty by using excisting filter systems.

3.3 Accident Analysis

In order to provide maximum safety of a fuel reprocessing plant, even under extreme conditions, all conceivable modes of maloperation and accidents are systematically analysed and precautions are taken to limit the consequences of such accidents to tolerable levels. The type of events analysed ranges from minor malfunctions, if relevant to plant safety, to a series of so-called design basis accidents, to which the plant must be able to cope with. With respect to the chemical processes and the materials handled in a reprocessing plant, the following main categories of possible accidents must be considered:

- chemical explosions
  (i.e. of a fission product or plutonium concentrator)
- fire
  (ignition of Zircaloy fines or burning of plutonium-loaded solvent)

- nuclear criticality
- loss of cooling accidents
  (i.e. in self-heating process or waste solutions)

In addition, a reprocessing plant, like other nuclear facilities, is designed for external impacts from man-made and natural catastrophes like

- plane crash
- shock wave from chemical explosion
- sabotage
- earthquake
- flood, storm etc.

3.3.2 Criticality accidents

The extent of a criticality accident in a geometrically non-safe vessel or equipment depends on the container dimensions and on the concentration of fissible and moderating material. The criticality excursion in a solution is an oscillating process:
Vapor bubbles, are formed, which are reducing the density and so the excursion is stopped for a short time. The expected fission rate will be not more than 10^{19} fissions during an accident of some minutes /19, 20, 21/.

In a reprocessing plant, all process equipment, as far as technically possible, is designed for inherent safety. In those parts of the plant, which are controlled by administrative safety measures the so-called multiple-contingency principle is applied.

Different criticality safety principles are applied for single process steps, i.e. homogeneous poisoning, heterogeneous poisoning and safety geometry. Criticality restrictions can be abandoned only within the Uranium purification cycles with uranyl-nitrate-solutions containing less than 1,8 % U-235.

A nuclear excursion is indicated by a suitable burst-alarm system. The thickness of the process cell walls guarantees a sufficient attenuation of gamma- and neutron radiation intensity and shields the operating personal. The off-gas cleaning system retains most of the activity carried by the formed vapor. So the permissible radiation dose values in the surroundings will not be exceeded.

3.3.2 Waste or Plutonium concentrator explosion accident

In the Savannah River Lab. 1953 an evaporator which contained concentrated uranyl nitrate, nitric acid and tributyl phosphate at a temperature of more than 160 ^{o}C exploded /22/. It was found out, that small amounts of solvent had left the extraction with the aqueous stream, were accumulated in the evaporator and built up to nitrated degradation products (red oil), which can react violently at temperatures above 150 ^{o}C.

To preclude such conditions it is now customary to wash the aqueous phase TBP-traces before entering a concentrator. In addition, the evaporator temperature is limited to 135 ^{o}C.

Assuming that both precautions would fail and reaction of red oil could librate an explosive energy of an equivalent amount of 2,5 kg TNT /23/ and destroy the evaporator. However, the design of the cell, layout of the ventilation ducts, and safety distance of filters will assure sufficient attenuation to maintain the integrity of containment systems and to limit the radiological risks to the immediate surrounding.

3.3.3 Ignition of Zircaloy Fines

The chopper of the fuel elements also produces very small zircaloy fines. Even it never occured in a reprocessing plant zirconium powder can inflame spontaneously. It is especially inflammable with a moisture content of 5 to 10 % /24/.

The zircaloy fines are separated out of the dissolver solution by a filter or a centrifuge. If the retention dries out an ignition could start and the fumes will carry fission product and plutonium particles.

Besides an eventual extension of the fire the worst consequences of this accident would consist in the release of radioactivity. Therefore the cell- and vessel-off gas systems have to be designed and equipped adequately.

3.3.4 Loss of cooling accident for self-heating solutions

Concentrated fission-product-solutions with high specific activities are self-heating and must be cooled during intermediate storage.The heat energy ranges up to 20 W/l. The loss of cooling will be considered as a design basis accident.
Stainless steel vessels are used for intermediate storage. They are equipped with jackets and internal coils for cooling. The cooling system consists of several redundant and independent circuits. The vessels as well as the auxiliary equipment are located process cells. These cells are designed to withstand all external impacts.

It must be demonstrated, that the cooling of the storage-vessels will be secured under the conditions of any external impacts or operational malfunction.

An extension of the accident analysis could show that the development of further safety systems will be required.

4. SAFETY ASPECTS OF MIXED OXIDE FUEL FABRICATION PLANTS

4.1 Potential hazard

Comparing the quantities of radioactive material (in Ci/a) in each plant of the fuel cycle in balance of considering the half-live of the essential plutonium isotopes, the sum of the actinides (Np, Pu, Am, Cm) indeed has the highest potential risk.

But potential risk and real hazard is not the same. Due to the drastic safety measures for the manufactoring of Pu in a Mixed Oxide Fuel Fabrication Plant (MOFFP) **the** risk for workers and the public is comparable with other industrial plants, e.g. of the chemical industry.

4.2 Normal operation

For the safe handlung of Pu in a MOFFP the greatest care is paid to the protection of the working staff against chronic or accidental radiation exposure by external radiation sources or intake of radioactive material and to avoid a dispersion and release of Pu-aerosols out of the plant. To ensure this protection the following safety measures are applied:

- The plutonium and plutonium containing material is strictly confined in tight glove-boxes, which are under constant underpressure.

- All glove-boxes are encircled by additional zones with also tight barriers.The plant exhaust systems provide for a stepwise decreasing of the air pressure in the direction of the areas with higher contamination risks, Fig. 5. The atmosphere in all zones is exchanged 2 - 8 times an hour.

- There are separate ventilation systems for both the glove-box-line and the building, Fig. 6. The exhausted air from the boxes and all working areas passed through several absolute filters connected in series before leaving the building. By these measures the amount of Pu which is released into the atmosphere with the ventilation air under normal operational conditions is negligible. In the ALKEM fabrication plant the released Pu in the past years did not reach 1 % of the maximum permissible value.

- The atmosphere inside the working rooms and all air streams of the plant are continuously monitored for contamination. In addition, to ensure an early detection of radioactivity dispersion all equipment and hands, feet and clothing of each worker are permanently checked.

- Where higher dose rate by external radiation must be expected for instance in the conversion and powder preparation area, additional shielding were installed. Besides the ALKEM production line has been built up in a way, that most of the process steps are mechanized and partly automized. The manual working inside of the glove-boxes by the staff is minimized as far as possible. The radiation hazard depends on the isotopic composition of the Pu and will increase with higher burn up of the fuel and repeated recycling of the Pu. So, for the future a further automation and especially a further limitation of manual glove-box operations to repair and maintenance work is to be expected.

- To avoid criticality accidents the quantity of fissile material handled has to be strictly controlled. Special attention must be devoted to abcut an undue accumulation of fissile material for example in the transport systems. As much as possible the nuclear safety is achieved by using the proper geometry. In addition a criticality alarm system is installed to assure immediately detection of a criticality event.

- To minimize the risks of fire and explosion the use of burnable or inflammable materials is kept as low as possible and strictly regulated. All areas of the plant are permanently supervised by means of fire detectors connected to an automatic extinguishing system for particular endangered areas.

4.3 Design basis accidents

From all accidents considered in a MOFFP the most serious would be an fire- or explosion-accident or criticality.

The effect of a fire or an explosion occuring in a glove-box or in working shop are experimentally defined. The damage would be locally limited and a severe contamination in this area is to be expected. But taking into account the above mentioned protection rules and measures there could not be a release of radioactivity out of the plant and consequences for the environment.

Several criticality accidents occured till now in nuclear plants manufactoring Uranium or Plutonium. In a criticality accident a maximum or 10^{18} fission can be produced. There are two kinds of risks for the environment.

The first is the exposure by direct gamma and neutron radiation emitted during the burst. But taking into account the shielding effect of walls and equipment of the plant the dangerous area is limited to a radius of several 10 meters around the point of event. The second risk consists in the gamma exposure emitted by the fission product. The radioactive cloud can result to a body burden of up approximately to 2 rem between 200 and 500 m. But in the case of a criticality too there will be no dispersion of Pu to the environment.

Another type of design basis accidents are events, which occur outside the plant but with consequences for the plant and therefore for the environment itself, like natural phenomena or plane-crash.

The probability of these events is very low. It is evaluated to be 10^{-6} - 10^{-7}/year for a plane-crash. To estimate the consequences of such a plane-crash onto a plutonium fabricating plant many factors are to be taken into account. Only few of these factors are well founded and hence the evaluations about Pu-release and radiological risks of the people at the site differ in a wide range. Although for the present there is an effort to minimize the risks of earthquakes and missiles by proper building constructions or limitation of the material quantities in a nuclear plant.

5. SUMMARY

Operational experience and detailed evaluation of the environmental impact during normal operation of LWR fuel reprocessing and mixed oxide fabrication plants lead to the conclusion that unavoidable adverse effects are small and within the range of variation of the natural background. The outstanding success of the "defense in depth" concept with commercial power reactors demonstrates that the safety approach applied in nuclear energy is indeed effective in protecting public health and safety.

The potential radiological hazard to the public involved in a large reprocessing plant is in the range of a large LWR. However, the following main differences are important from a safety point of view:

- Both fuel reprocessing and fabrication plants do not have the high system pressure that is associated with light water reactors.

- In both fuel reprocessing and fabrication plants a stepwise decrease of system pressures is provided in the direction of zones with increasing radioactive inventories.
- A considerable amount of the radioactivity of the fuel which is in the form of short lived radionuclides has decayed before reprocessing.
- Most of the fission product inventory accumulated in a reprocessing plant (except for the fuel storage pool) is in the liquid state, in constrast to reactor conditions.
- In spite of the degree of automation already achieved, Plutonium fuel manufacturing still implies the largest fraction of direct human access to pure fissionable material in the whole fuel cycle.

Realistic or best estimate analysis of design basis accidents for a large reprocessing plant have shown that the associated activity releases would result in much lower doses than the maximum doses established in the regulatory criteria. These criteria can be kept too, if extremely pessimistic assumptions are made.

The measures to further mitigate adverse effects of both normal operation and postulated accidents are under development. This development includes in particular highly efficient retention systems for Plutonium and Transuranium isotopes, Krypton 85, Iodine 129, Tritium to be applied in large reprocessing plants.

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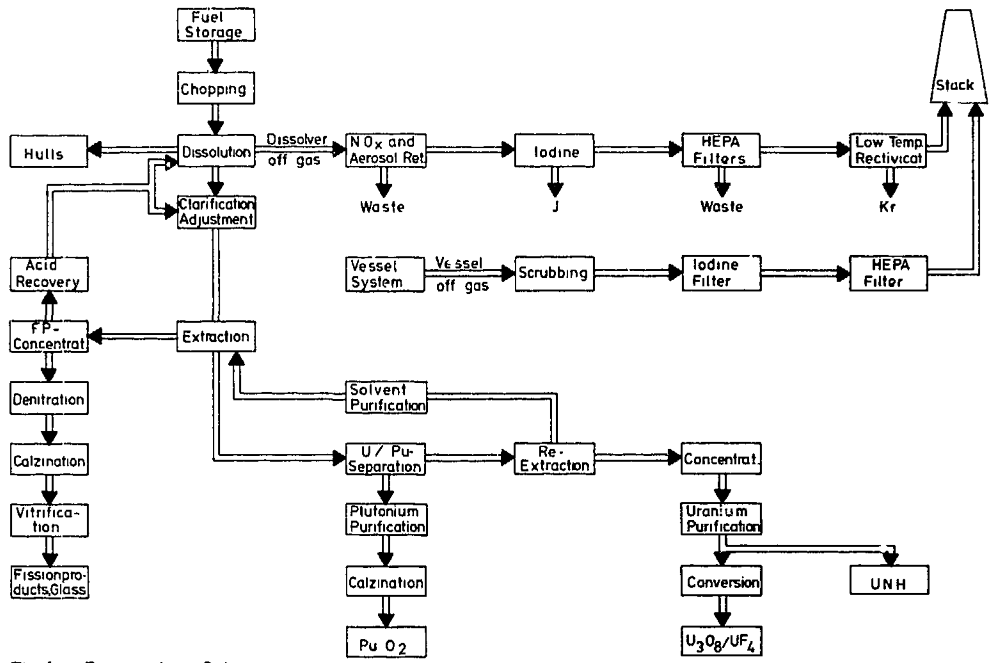

Fig.1 Processing Scheme

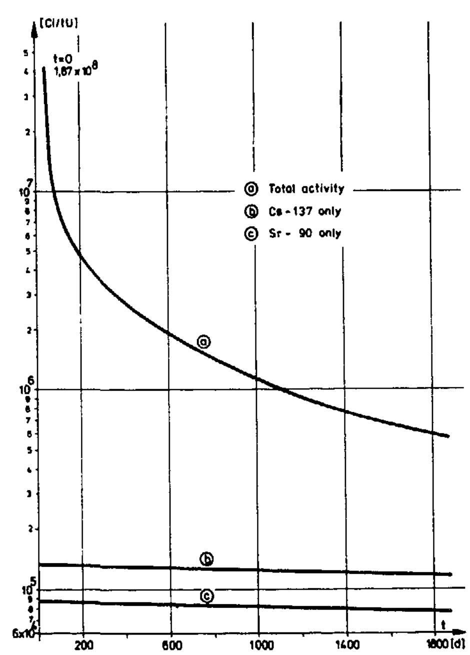

Fig. 2: Activity / tU as function of the fuel element cooling time

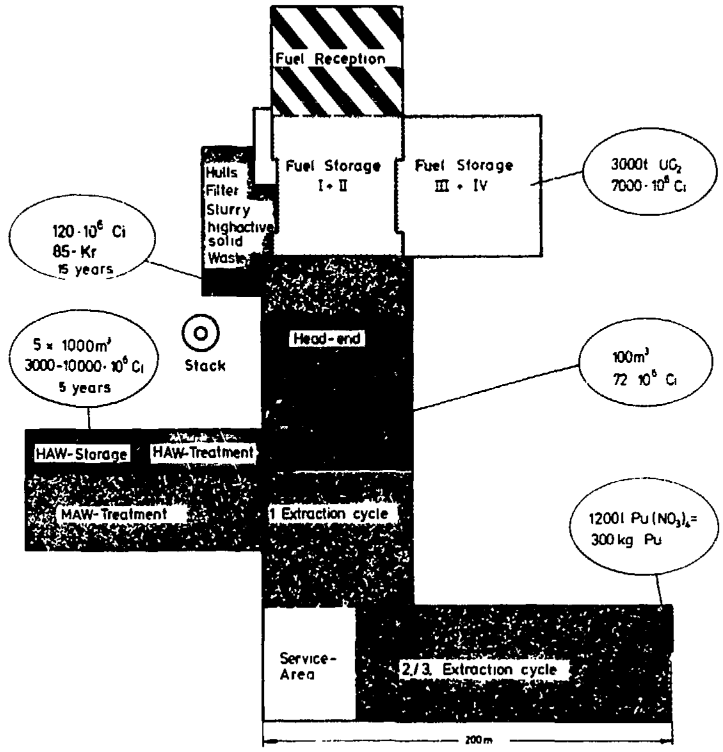

Fig.3: Lay-out of the Buildings and Activity Inventory of a Reprocessing Plant with a Capacity of 1500 t/a

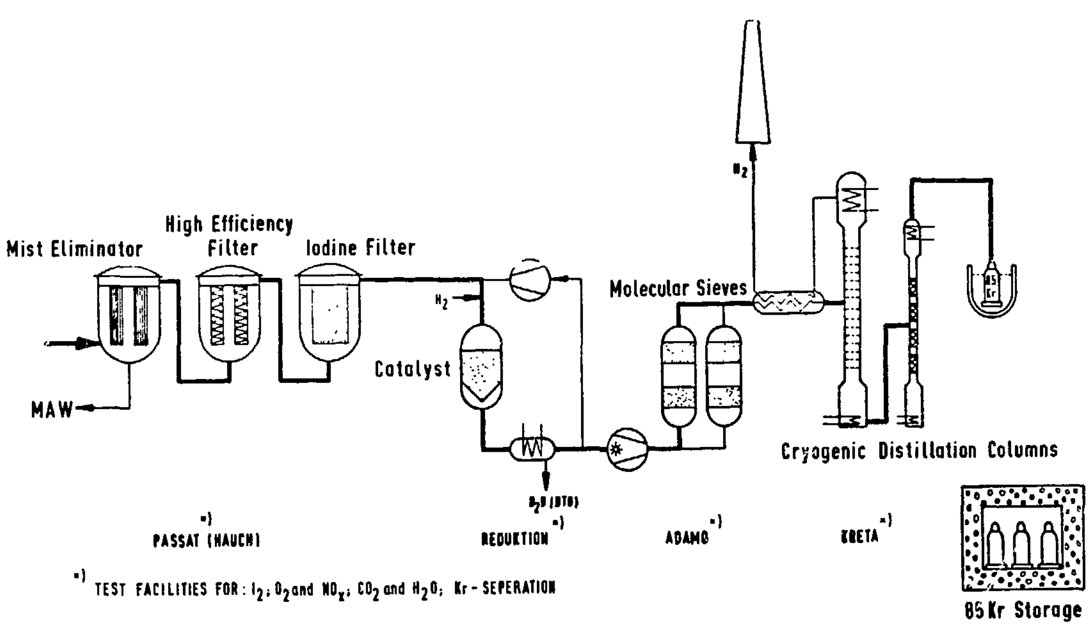

Fig. 4: Dissolver Offgas Cleaning System of a Reprocessing Plant

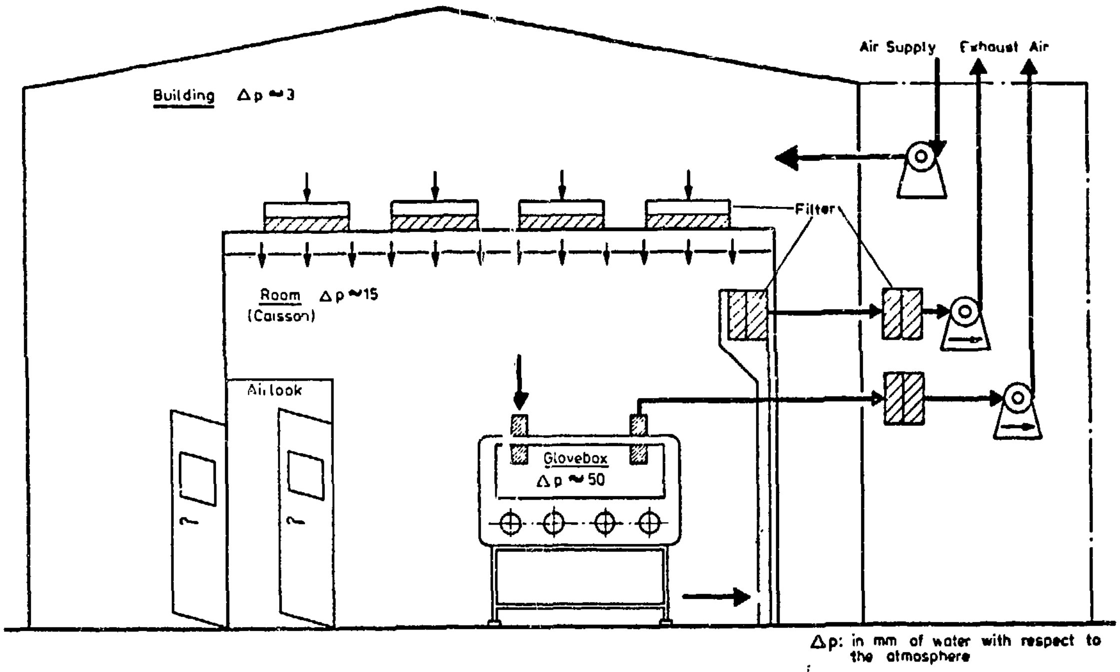

Fig. 5: Sketch of the Multiple Containments with Negative Pressure Zones

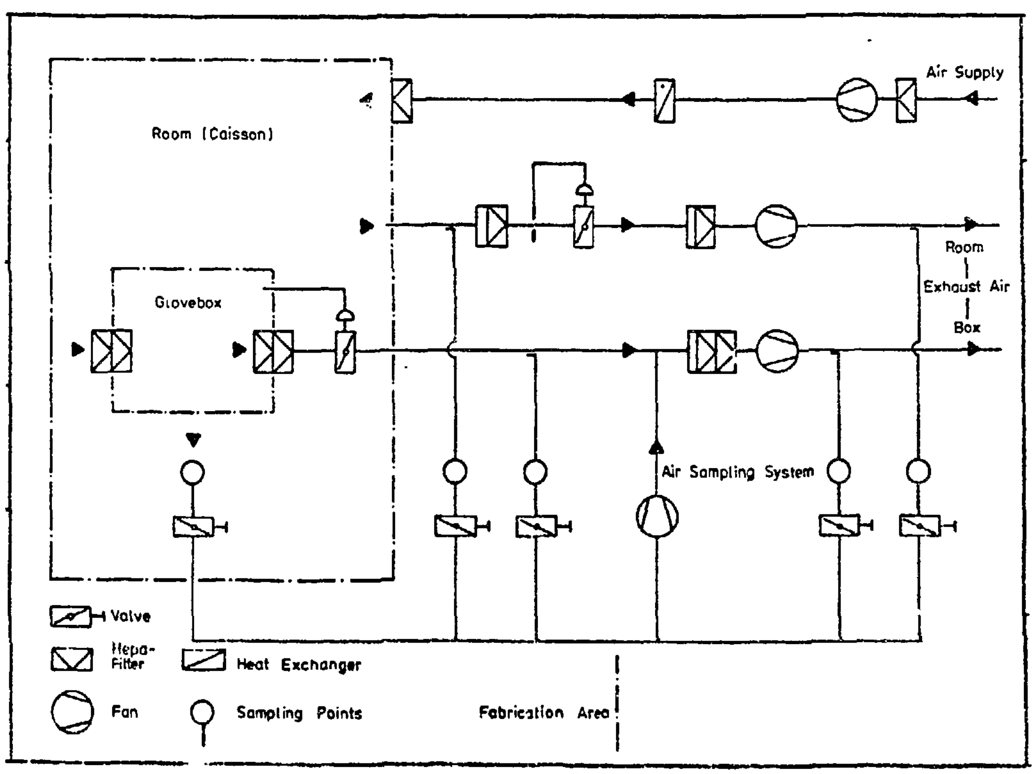

Fig. 6: Schematic Diagram of the Ventilation System